JC17 Rec'd PCT/PTO 25 MAY 2001

FORM PTO-1390 (REV 11-2000)	U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER **1721-30**
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (If known, see 37 C.F.R. 1.5) 09/856710 Unknown
INTERNATIONAL APPLICATION NO. **PCT/FR98/02563**	INTERNATIONAL FILING DATE **27 November 1998**	PRIORITY DATE CLAIMED **27 November 1997**

TITLE OF INVENTION
MEANS FOR GENERATING OPTICAL RADIATIONS TUNEABLE AT LEAST IN FREQUENCY

APPLICANT(S) FOR DO/EO/US
BOULANGER et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The U.S. has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☐ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. ☒ is attached hereto (required only if not communicated by the International Bureau).
   b. ☒ has been communicated by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. ☒ is attached hereto.
   b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are attached hereto (required only if not communicated by the International Bureau).
   b. ☐ have been communicated by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has **NOT** expired.
   d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☒ A English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 To 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information. PTO-1449/ International Search Report/ Front page of the PCT Publication/ Translator's Certificate; a Request pursuant to Rule 137(b)

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U.S. APPLICATION NO (If known, see 37 C.F.R. 1.5)	INTERNATIONAL APPLICATION NO	ATTORNEY'S DOCKET NUMBER
Unknown 09/856710	PCT/FR98/02563	1721-30

21. ☒ The following fees are submitted: — CALCULATIONS — PTO USE ONLY

BASIC NATIONAL FEE (37 C.F.R. 1.492(a)(1)-(5):

- -- Neither international preliminary examination fee (37 C.F.R. 1.482) nor international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO $1000.00
- -- International preliminary examination fee (37 C.F.R. 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO....................$860.00
- -- International preliminary examination fee (37 C.F.R. 1.482) not paid to USPTO but international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO........................$710.00
- -- International preliminary examination fee (37 C.F.R. 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)...............................$690.00
- -- International preliminary examination fee (37 C.F.R. 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4).......................................$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =						$	860.00
Rule 17(m) fee						$	1240.00
CLAIMS	NUMBER FILED		NUMBER EXTRA	RATE			
Total Claims	39	-20 =	19	X	$18.00	$	342.00
Independent Claims	1	-3 =	0	X	$80.00		0.00
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)				$270.00		$	0.00
TOTAL OF ABOVE CALCULATIONS =						$	**2442.00**
☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.							0.00
SUBTOTAL =						$	**2442.00**
Processing fee of $130.00, for furnishing the English Translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. 1.492(f)). +							0.00
TOTAL NATIONAL FEE =						$	**2442.00**
Fee for recording the enclosed assignment (37 C.F.R. 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31). **$40.00** per property +						$	0.00
Fee for Petition to Revive Unintentionally Abandoned Application ($1240.00 – Small Entity = $620.00)						$	0.00
TOTAL FEES ENCLOSED =						$	**2442.00**
						Amount to be: refunded	$
						Charged	$

a. ☒ A check in the amount of $2442.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. 14-1140 in the amount of $_____ to cover the above fees. A duplicate copy of this form is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 14-1140. A duplicate copy of this form is enclosed.

d. ☒ The entire content of the foreign application(s), referred to in this application is/are hereby incorporated by reference in this application.

NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

B. J. Sadoff
NAME

36,663 — **May 25, 2001**
REGISTRATION NUMBER — Date

528517

09/856710
JC18 Rec'd PCT/PTO 25 MAY 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

BOULANGER et al. — Atty. Ref.: 1721-30

U.S. National Phase of PCT/FR98/02563 — Group:

Serial No. Unknown

Filed: May 25, 2001 — Examiner:

For: MEANS FOR GENERATING OPTICAL RADIATIONS TUNEABLE AT LEAST IN FREQUENCY

* * * * * * * * * * *

May 25, 2001

Assistant Commissioner for Patents
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

Preliminarily amend the above-identified application as follows:

IN THE CLAIMS

Amend the claims as follows:

3. (Amended) The device according to claim 1, characterized in that said cylindrical volume has a section selected from a circular section and an elliptical section.

4. (Amended) The device according to claim 1, characterized in that said crystal includes at least one hyperpolarizable chemical entity.

5. (Amended) The device according to claim 1, characterized in that said crystal is a crystal selected from a crystal of $LiTaO_3$, $KTiOPO_4$, $KTiOAsO_4$, $RbTiOPO_4$, $RbTiOAsO_4$, $CsTiOAsO_4$, $\beta.Ba_4BO_4$, LiB_3O_5, $KNbO_3$, $LiIO_3$, $LiNbO_3$, KD_2PO_4, $KH_2P\ O_4$, $NH_4H_2PO_4$, CsD_2ASO_4, CsH_2AsO_4, $AgGaS_2$, $AgGAsO_2$, $ZnGeP_2$, Tl_3AsSe_3 and a crystal of GaAs.

6. (Amended) The device according to claim 1, characterized in that said optical system is essentially formed by two components placed on both sides of said crystal and selected from a convergent lens, a divergent lens, a set of lenses, a reflecting surface or mirror with the concavity orientated on the side of said crystal, and a reflecting surface or mirror with the concavity orientated on the opposite side of said crystal.

7. (Amended) The device according to claim 1, characterized in that the axis of revolution of said crystal coincides with a rotary mechanical axis so that the crystal may rotate around its axis.

8. (Amended) The device according to claim 1, characterized in that said crystal is a crystal with a phase tuning property through double refraction.

10. (Amended) The device according to claim 1, characterized in that said crystal is a crystal with a quasi phase tuning property.

12. (Amended) The device according to claim 1, characterized in that the axis of revolution of said crystal is orthogonal to the plane of the direction(s) of wave vectors of a sought-after interaction, and more particularly of the direction(s) providing maximum efficiency of this interaction.

13. (Amended) The device according to claim 1, characterized in that said crystal contains direction(s) providing a maximum efficiency of the interaction, so that they are accessible to said incident optical radiation(s) under normal incidence or close to the normal on one or more surface(s) of said crystal defining a cylindrical volume of revolution, either by rotation of said crystal around its axis of revolution, or by rotation of said incident optical radiations around said crystal in a plane orthogonal to the axis of revolution of said crystal.

14. (Amended) The device according to claim 1, characterized in that said incident optical radiation(s) comprise (each) one, two, three or four equal or different frequencies, with colinear or non-colinear wave vectors, and under normal incidence or close to the normal on one or more surfaces of said crystal defining a cylindrical volume of revolution.

15. (Amended) The device according to claim 1, characterized in that said crystal has a network of monocrystalline domains selected from a network of plane monocrystalline domains, a network of circular monocrystalline domains, a network of elliptical monocrystalline domains.

16. (Amended) The device according to claim 1, characterized in that said crystal has a network of periodically alternating domains, optionally surrounded by a non-alternating monocrystalline crown (c).

17. (Amended) The device according to claim 1, characterized in that said incident optical radiations are laser radiation(s), notably one or more laser radiations selected from a fixed frequency laser radiation and a tunable frequency laser radiation.

18. (Amended) The device according to claim 1, characterized in that said interaction(s) are interactions (s) selected from a three-wave interaction or a four-wave interaction.

19. (Amended) The device according to claim 1, characterized in that said crystal has a non-centrosymmetric structure so that said device provides a three-wave interaction.

20. (Amended) The device according to claim 1, characterized in that said or at least one of said incident optical radiation(s) comprises two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to the sum of said two, or, if required, said three frequencies comprised in said incident optical radiation(s).

21. (Amended) The device according to claim 1, characterized in that said or at least one of said emerging optical radiation(s) comprise a frequency which is equal to the double or the triple of a frequency comprised in said or at least one of said incident optical radiation(s).

22. (Amended) The device according to claim 1, characterized in that said or at least one of said incident optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to a difference between said two, or if required, said three frequencies comprised in said incident optical radiation(s).

23. (Amended) The device according to claim 1, characterized in that said or at least one of said emerging optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, the sum of which is equal to a frequency comprised in said or at least one of said incident optical radiation(s).

24. (Amended) The device according to claim 1, characterized in that said or at least one of said interaction(s) is an interaction with colinear wave vectors.

25. (Amended) The device according to claim 1, characterized in that said interaction is an interaction with non-colinear wave vectors.

26. (Amended) The device according to claim 1, characterized in that said or at least one of said interaction(s) is an interaction selected from an optical parametric amplification, a generation of second or third harmonic.

27. (Amended) The device according to claim 1, characterized in that said crystal is placed inside a cavity providing a resonant interaction, and in that said optical system for confining and focussing said incident optical radiation(s) on the central portion(s) of said crystal on the one hand, and for collimating and directing said emerging optical radiation(s) on the other hand, is placed outside said cavity.

29. (Amended) The device according to claim 27, characterized in that said cavity includes input and output reflecting surfaces facing each other providing resonance for at least one of the interacting waves.

30. (Amended) The device according to claim 29, characterized in that said input reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

31. (Amended) The device according to claim 29, characterized in that said at least one resonant wave has a non-zero double refraction angle ρ and in that said output reflecting surface has a radius of curvature, with a concavity selected from a concavity orientated on the side of said

crystal and a concavity orientated on the opposite side of said crystal, so that the outgoing and returning beams coincide.

32. (Amended) The device according to claim 29, characterized in that said at least one resonant wave has a non-zero double refraction angle ρ, and in that said output reflecting surface is placed at a distance (d) from said crystal and has a radius of curvature R the respective values of which satisfy equation $R = d - L$ with d larger than L for a concavity orientated on the side of said crystal, or the equation $R = L - d$ with d less than L for a concavity orientated on the opposite side of said crystal, with L defined as $L = R_c (\cos(2\rho) + (\sin(2\rho) / \tan(\rho_e)) - 1)$, with R_c the radius of the cylindrical volume of revolution, ρ the double refraction angle and with ρ_e defined by $\rho_e = \arcsin(n \sin(2\rho) - 2\rho)$, with n being the refractive index of said at least one wave for which resonance is sought.

33. (Amended) The device according to claim 29, characterized in that said at least one resonant wave has a zero double refraction angle ρ, and in that said output reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

34. (Amended) The device according to claim 1, characterized in that it further comprises means for thermostatic control of said crystal.

35. (Amended) The device according to claim 1, characterized in that said crystal is held at a temperature different from room temperature.

36. (Amended) The device according to claim 1, characterized in that it further comprises means for applying a static or low frequency electric field to the inside of said crystal.

37. (Amended) The device according to claim 1, characterized in it further comprises a pair of electrodes placed on the opposite faces of said crystal.

38. (Amended) The device according to claim 1, characterized in that it forms a component selected from a spectroscope component, a remote detection system component, a remote

transmission system component, a remote guiding system component, a LIDAR system component, an optronic counter-measure system component.

39. (Amended) A method for generating an optical radiation at least tunable in frequency, characterized in that it implements a device according to claim 1.

REMARKS

Entry of the above is requested to reduce improper multiple dependencies.

Respectfully submitted,

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By: ______________________

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS

3. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said cylindrical volume has a section selected from a circular section and an elliptical section.

4. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal includes at least one hyperpolarizable chemical entity.

5. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal is a crystal selected from a crystal of $LiTaO_3$, $KTiOPO_4$, $KTiOAsO_4$, $RbTiOPO_4$, $RbTiOAsO_4$, $CsTiOAsO_4$, $\beta.Ba_4BO_4$, LiB_3O_5, $KNbO_3$, $LiIO_3$, $LiNbO_3$, KD_2PO_4, $KH_2P\ O_4$, $NH_4H_2PO_4$, CsD_2ASO_4, CsH_2AsO_4, $AgGaS_2$, $AgGAsO_2$, $ZnGeP_2$, Tl_3AsSe_3 and a crystal of GaAs.

6. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said optical system is essentially formed by two components placed on both sides of said crystal and selected from a convergent lens, a divergent lens, a set of lenses, a reflecting surface or mirror with the concavity orientated on the side of said crystal, and a reflecting surface or mirror with the concavity orientated on the opposite side of said crystal.

7. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that the axis of revolution of said crystal coincides with a rotary mechanical axis so that the crystal may rotate around its axis.

8. (Amended) The device according to [any of claims 1 to 7] <u>claim 1</u>, characterized in that said crystal is a crystal with a phase tuning property through double refraction.

10. (Amended) The device according to [any of claims 1 to 7] <u>claim 1</u>, characterized in that said crystal is a crystal with a quasi phase tuning property.

12. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that the axis of revolution of said crystal is orthogonal to the plane of the direction(s) of wave

vectors of a sought-after interaction, and more particularly of the direction(s) providing maximum efficiency of this interaction.

13. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal contains direction(s) providing a maximum efficiency of the interaction, so that they are accessible to said incident optical radiation(s) under normal incidence or close to the normal on one or more surface(s) of said crystal defining a cylindrical volume of revolution, either by rotation of said crystal around its axis of revolution, or by rotation of said incident optical radiations around said crystal in a plane orthogonal to the axis of revolution of said crystal.

14. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said incident optical radiation(s) comprise (each) one, two, three or four equal or different frequencies, with colinear or non-colinear wave vectors, and under normal incidence or close to the normal on one or more surfaces of said crystal defining a cylindrical volume of revolution.

15. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal has a network of monocrystalline domains selected from a network of plane monocrystalline domains, a network of circular monocrystalline domains, a network of elliptical monocrystalline domains.

16. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal has a network of periodically alternating domains, optionally surrounded by a non-alternating monocrystalline crown (c).

17. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said incident optical radiations are laser radiation(s), notably one or more laser radiations selected from a fixed frequency laser radiation and a tunable frequency laser radiation.

18. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said interaction(s) are interactions (s) selected from a three-wave interaction or a four-wave interaction.

19. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said crystal has a non-centrosymmetric structure so that said device provides a three-wave interaction.

20. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said or at least one of said incident optical radiation(s) comprises two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to the sum of said two, or, if required, said three frequencies comprised in said incident optical radiation(s).

21. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said or at least one of said emerging optical radiation(s) comprise a frequency which is equal to the double or the triple of a frequency comprised in said or at least one of said incident optical radiation(s).

22. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said or at least one of said incident optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to a difference between said two, or if required, said three frequencies comprised in said incident optical radiation(s).

23. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said or at least one of said emerging optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, the sum of which is equal to a frequency comprised in said or at least one of said incident optical radiation(s).

24. (Amended) The device according to [any of the claims 1 to 23] claim 1, characterized in that said or at least one of said interaction(s) is an interaction with colinear wave vectors.

25. (Amended) The device according to [any of claims 1 to 24] claim 1, characterized in that said interaction is an interaction with non-colinear wave vectors.

26. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said or at least one of said interaction(s) is an interaction selected from an optical parametric amplification, a generation of second or third harmonic.

27. (Amended) The device according to [any of the preceding claims] claim 1, characterized in that said crystal is placed inside a cavity providing a resonant interaction, and in that said optical system for confining and focussing said incident optical radiation(s) on the central portion(s) of said crystal on the one hand, and for collimating and directing said emerging optical radiation(s) on the other hand, is placed outside said cavity.

29. (Amended) The device according to claim 27 [or 28], characterized in that said cavity includes input and output reflecting surfaces facing each other providing resonance for at least one of the interacting waves.

30. (Amended) The device according to [any] claim 29, characterized in that said input reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

31. (Amended) The device according to claim 29 [or 30], characterized in that said at least one resonant wave has a non-zero double refraction angle ρ and in that said output reflecting surface has a radius of curvature, with a concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side of said crystal, so that the outgoing and returning beams coincide.

32. (Amended) The device according to [any of the claims 29 to 31] claim 29, characterized in that said at least one resonant wave has a non-zero double refraction angle ρ, and in that said output reflecting surface is placed at a distance (d) from said crystal and has a radius of curvature R the respective values of which satisfy equation $R = d - L$ with d larger than L for a concavity orientated on the side of said crystal, or the equation $R = L - d$ with d less than L for a concavity orientated on the opposite side of said crystal, with L defined as $L = R_c (\cos(2\rho) + (\sin(2\rho) / \tan(\rho_e)) - 1)$, with R_c the radius of the cylindrical volume of revolution, ρ the double refraction

angle and with ρ_e defined by $\rho_e = \arcsin(n \sin(2\rho) - 2\rho)$, with n being the refractive index of said at least one wave for which resonance is sought.

33. (Amended) The device according to claim 29 [or 30], characterized in that said at least one resonant wave has a zero double refraction angle ρ, and in that said output reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

34. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that it further comprises means for thermostatic control of said crystal.

35. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that said crystal is held at a temperature different from room temperature.

36. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that it further comprises means for applying a static or low frequency electric field to the inside of said crystal.

37. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in it further comprises a pair of electrodes placed on the opposite faces of said crystal.

38. (Amended) The device according to [any of the preceding claims] <u>claim 1</u>, characterized in that it forms a component selected from a spectroscope component, a remote detection system component, a remote transmission system component, a remote guiding system component, a LIDAR system component, an optronic counter-measure system component.

39. (Amended) A method for generating an optical radiation at least tunable in frequency, characterized in that it implements a device according to [any of the preceding claims] <u>claim 1</u>.

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(81) Etats désignés: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, brevet ARIPO (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), brevet eurasien (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), brevet européen (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), brevet OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

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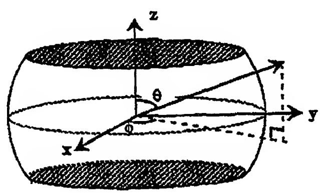
(54) Title: MEANS FOR GENERATING OPTICAL RADIATIONS TUNEABLE AT LEAST IN FREQUENCY

(54) Titre: MOYENS POUR LA GENERATION DE RAYONNEMENTS OPTIQUES ACCORDABLES AU MOINS EN FREQUENCE

(57) Abstract

The invention concerns a device for generating, by interaction(s) with three or four waves from one or several incident optical radiation(s), one or several emergent radiation(s) tuneable at least in frequency. The invention is characterised in that it consists essentially of a crystal with non-linear optical property whereof the surface defines a cylindrical volume with complete revolution, or truncated on at least two opposite faces symmetrical relative to its axis of revolution, or partial on one of two said faces. Said device is particularly designed for applications in spectroscopy, remote sensing, long-distance transmission, remote guiding. The invention also concerns a method using said device.



cylindroïde 2

cylindroïd 2

(57) Abrégé

La présente invention concerne un dispositif pour la génération, par interaction(s) à trois ou quatre ondes à partir d'un ou plusieurs rayonnement(s) optique(s) incident(s), d'un ou plusieurs rayonnement(s) optique(s) émergent(s) accordable(s) au moins en fréquence. Le dispositif selon l'invention est essentiellement constitué par un cristal à propriété optique non linéaire dont la surface définit un volume cylindrique de révolution de manière complète, ou bien de manière tronquée sur au moins deux cadrans opposés et symétriques par rapport à son axe de révolution, ou bien de manière partielle sur un seul de tels cadrans. Le dispositif selon l'invention est particulièrement destiné à des applications de spectroscopie, de télédétection, de télétransmission, de téléguidage. La présente invention concerne également une méthode mettant en oeuvre un tel dispositif.

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MEANS FOR GENERATING OPTICAL RADIATIONS AT LEAST TUNABLE IN FREQUENCY

The present invention generally relates to a device and a method for generating optical radiations at least tunable in frequency.

Various devices and methods are presently available for frequency conversion by non-linear optical interaction with phase tuning or quasi phase tuning. However none of these devices is fully functional and/or satisfactory.

On this account, the example of devices using parallelepiped-shaped monocrystals may be mentioned. Certain of these parallelepipedal monocrystal devices are for frequency conversion by non-linear optical interaction with phase tuning through double refraction. The orientation of the faces of the parallelepiped is then selected according to the desired interaction. Thus, in most cases, as for the generation of the second harmonic for example, a crystal may only be used for a given interaction, i.e.,

for a particular doublet of frequency (v, 2v), the associated phase tuning direction is orthogonal to the polished faces. However, there are devices based on parallelepipedal crystals which make use of the angular tunability for parametric amplification or oscillation for example. Because of the losses induced by refraction and the non-colinearity of wave vectors under oblique incidence, there is only a small angular range of the crystal which may be used, typically about ten degrees of external angle on both sides of the directions orthogonal to both parallel faces subject to the radiation. Such an angular deviation is not always sufficient for accessing the entire existing phase tuning directions. On the other hand, refraction under oblique incidence leads to deformation and spectral widening of the generated beams.

Other tunable devices with parallelepipedal materials are for frequency conversion by non-linear optical interaction with quasi phase tuning. They now use translation with respect to the radiation of a parallelepipedal sample in which several gratings with different periodicities have been engraved. The main drawback of this technique is that it is necessary to achieve, for each jump from one grating to the other, angular or thermal tuning in order to make the generated radiation's spectral band continuous.

This is valid whether the interaction is resonant or not, i.e., whether the sample is placed in a cavity or not.

Other devices of the prior art require an index adaptation medium, and/or do not exhibit satisfactory spectral fineness on a large range of tunability.

The object of the present invention is to overcome the drawbacks of the devices and techniques of the prior art, and it provides a device for generating, through three- or four-wave interaction(s) from one or more incident optical radiation(s), one or more emerging optical radiation(s) at least tunable in frequency, characterized in that it essentially comprises a crystal with a non-linear optical property, the surface of which defines a cylindrical volume of revolution, in a complete or truncated way on at least two opposite and symmetrical quadrants with respect to its axis of revolution or else partly on a single one of these quadrants, and in that it further comprises an optical system for confining and focussing said incident optical radiation(s) on the central portion of said crystal on the one hand, and for collimating and directing said emerging optical radiation(s).

A cylindrical volume of revolution, in the present application, means a volume of revolution described by a line or curve segment moving along two closed curves located in parallel planes. These closed curves may be selected from circles and ellipses.

The term cylindrical volume of revolution therefore means a volume of a cylinder as illustrated under reference number 1 in Fig. 2 (volume described by a curve segment) and a volume of a cylindroid as

illustrated under reference number 2 in Fig. 2 (volume described by a curve segment). It should be emphasized that these cylindrical volumes of revolution (cylinder and cylindroid) may have a section selected from a circular section and an elliptical section. Such a section may notably be considered as following a plane orthogonal to the axis of revolution.

This cylindrical volume of revolution comprises, when it is described in a complete way, a basic volume, from which may be derived volumes described in a truncated way, or even in a partial way. Indeed, taking into account the symmetry of the index surface, a rotation of $\Delta\alpha = 90°$ of a crystal with a cylindrical volume of revolution with respect to a main axis of the index surface is sufficient for accessing the whole of the possible non-linear optical interactions for frequency conversion.

The cylindrical volume of revolution may therefore be only machined in a truncated way, or in a partial way, while maintaining access to the whole of the possible interactions. Of course, it may also be decided to only machine said cylindrical volume in a truncated or partial way for the simple purpose of limiting the machining to the subset of the particular sought-after interactions.

The truncated machining may only be performed, with respect to the cylindrical volume of revolution, on two opposite properly orientated cylindrical quadrants, symmetrical with respect to the rotation

axis, or even on two quadrant portions, the angular deviation $\Delta\alpha$ of which allows propagation of the various sought-after interactions. The resulting volume corresponds to a cylindrical volume of revolution described in a truncated way. Such a truncated volume is illustrated under reference number 3 (truncated cylinder) and 4 (truncated cylindroid) in Fig. 2.

Alternatively, only one of these quadrants or portions of quadrants may be machined. The resulting volume corresponds to a cylindrical volume of revolution described in a partial way. Such a partial volume is illustrated in Fig. 2 under reference number 5 (cylinder or cylindroid portion along planes containing the axis of revolution).

The crystal of the device according to the invention may thus notably have a volume selected from a cylinder volume 1, a cylindroid volume 2, a truncated cylinder volume 3 or 4 (having at least two opposite quadrants of cylindrical volume of revolution and symmetrical with respect to the axis of revolution of said crystal), a partial cylinder or cylindroid volume 5 (cylinder or cylindroid portion having only one of such quadrants). Said cylinder or cylindroid volumes may have a circular section or an elliptical section, notably along a plane orthogonal to the axis of revolution.

Said crystal has, at least on its useful surfaces, a surface condition suitable for achieving optical interactions. In particular, the surface(s) of said

crystal which define a cylindrical volume of revolution are optically polished.

The term "optical radiation", in the present application refers to a beam of electromagnetic waves with a frequency or frequencies belonging to the ultraviolet and/or visible and/or infrared spectrum. The value of this frequency or these frequencies is between about 1 and 15,000 nanometers, and more particularly between 100 and 10,000 nanometers.

A non-linear optical property means in the present application an optical frequency conversion property and/or an electro-optical property.

The device according to the invention has many advantages. Indeed, various types of interactions (notably, optical parametric amplification, generation of second or third harmonics, optical parametric oscillation) may be achieved with it, and this, with phase tuning as well as with quasi phase tuning. It may also be emphasized that, with the device according to the invention, three-wave interactions as well as four-wave interactions may be achieved, and it allows colinear wave vector interactions, in the same way as non-linear wave vector interactions.

The device according to the invention also has the advantage of being able, with a single crystal, to generate radiations with better spatial quality and better spectral fineness, and this over a larger spectral band as compared with presently available devices.

The device according to the invention actually provides refraction under normal or near normal incidence for an elliptical grating, with respect to the surfaces receiving the radiations, regardless of the orientation of the non-linear crystal with respect to the beams, as long as the beams propagate along a diameter of said cylindrical volume. The term "near the normal" aims at a direction with an angular deviation $i(\alpha)$ from the normal direction, such as defined in the following Example 12 and in Fig. 11. Because of this, the interaction for converting frequencies in the crystal always occurs with colinear wave vectors which allows beams to be generated with a better transverse energy profile and better spectral fineness as in the case of the prior art devices. Indeed, in the prior art devices, having a crystal with parallelepipedal geometry, the frequency conversion interaction generally occurs with non-colinear wave vectors as soon as the non-linear crystal is set into rotation and as soon as the beams are no longer under normal or quasi normal incidence, but under oblique incidence; the quality of the emitted beams thereby deteriorates and this all the more since non-colinearity is important.

The device according to the invention is adapted for (and specifically optimized for) tunability in frequency, it utilizes a collimating/focussing system, which notably allows it to control propagation of the radiations. The confining and collimating optical system of the device according to the invention notably

is able to provide an optimized control of the propagation of the radiations: it thus offers better spectral fineness associated with a wider tunability spectral range.

It also enables the crystal to be used without it being necessary to immerse it in an index-adapting medium such as a refractive index adapting liquid. Indeed, the device according to the invention does not need any index adapting medium: said crystal may be placed in free air, or in any other medium with an index. The device according to the invention therefore has also the advantages of mechanical simplicity (simplification of the system for holding and setting into rotation the non-linear crystal).

The device according to the invention is particularly adapted for crystals which include at least a hyperpolarizable chemical entity. Said crystal is preferably a crystal selected from a crystal of $KTiOPO_4$, $HTiOAsO_4$, $RbTiOPO_4$, $RbTiOAsO_4$, $CsTiOAsO_4$, $\beta.BaB_2O_4$, LiB_3O_5, $KNbO_3$, $LiIO_3$, $LiNbO_3$, $LiTaO_3$, KD_2PO_4, KH_2PO_4, $NH_4H_2PO_4$, CsD-AsO_4, CsH_2AsO_4, $AgGaS_2$, $AgGaSe_2$, $ZnGeP_2$, Tl_3AsSe_3 and a crystal of GaAs. Advantageously, the size of said crystal is selected from a micrometric size, a millimetric size and a centimetric size.

According to a configuration of this advantageous aspect, said optical system essentially comprises at least two components placed on both sides of said crystal and selected from a convergent lens, a set of lenses, a reflecting surface or a mirror with the

concavity orientated on the side of said crystal, and a reflecting surface or a mirror with the concavity orientated on the opposite side of said crystal. They are selected in order to have a focus and a geometry such that they may be placed at such a focal distance that a first one of them is capable of focussing laser radiation incident on said crystal, and that a second one of them is capable of reducing the divergence of the laser radiation emerging from the crystal. The device according to the invention may comprise one or more pair(s) of such optical components and in particular one or more pair(s) of lenses.

The focal distance of said components is such that the radiation(s) have a dimension which is small as compared to the radius of said crystal in order to limit optical aberrations and to increase the interaction efficiency, and such that this dimension is sufficient in order to prevent any damage to the crystal by a too strong incident intensity. This focal distance is generally between about 50 and 500 mm.

According to another advantageous aspect of the invention, the axis of revolution of said crystal coincides with a rotary mechanical axis so that the crystal may turn around its axis. Such a crystal may then be securely fixed to a goniometric device, advantageously, for controlling its angular displacement.

According to still another advantageous aspect of the invention, said crystal is a crystal with the

property of phase tuning through double refraction, such as a monocrystal, or else it is a crystal with a property of quasi phase tuning.

A so-called crystal with a property of quasi phase tuning may have along the sought-after direction of propagation of the radiations, a periodically alternating juxtaposition of monocrystalline domains, i.e., a juxtaposition of monocrystalline layers turned by an angle of 180°, one with respect to the other. Such a crystal with the property of quasi phase tuning may also be obtained through machining and juxtaposition of monocrystals, or else even by applying an electric field to a grid of electrodes deposited on the faces orthogonal to the polar axis of a ferroelectric crystal. Advantageously a crystal results, the network of which has a periodicity vector orthogonal to the axis of revolution of said crystal, and which has an effective non-linear coefficient with a periodically alternating sign according to intervals depending on the relevant direction of propagation. The interaction for which the coherence length is equal to an odd multiple of the monocrystalline domain width in the relevant direction of propagation has therefore a maximum efficiency. Alternatively, a crystal with the property of quasi phase tuning adapted to the device according to the invention may have different values for the refractive index, modulated in a periodical way according to the relevant direction of propagation.

According to a further advantageous aspect of the

invention, the axis of revolution of said crystal is orthogonal to the plane of the direction(s) of wave vectors of an sought-after interaction, and more particularly to that of the direction(s) providing maximum efficiency for this interaction.

According to a particularly advantageous aspect of the invention, said crystal contains the direction(s) providing maximum efficiency for the interaction, so that they are accessible to said incident optical radiation(s) under normal or near normal incidence (deviation by an angle $i(\alpha)$ with respect to the normal, as defined in the following Example 12 - formula 21, notably, and in Fig. 11), with respect to the surfaces receiving the radiations, on one of the surface(s) of said crystal defining a cylindrical volume of revolution, either by rotation of said crystal around its axis of revolution, or by rotation of said incident optical radiation(s) around said crystal in a plane orthogonal to the axis of revolution of said crystal. This particularly advantageous aspect of the invention in the case of said crystal with the property of phase tuning by double refraction, enables only one crystal to be used instead of a plurality of parallelepipedal crystals, and in the case of said crystal with the property of quasi phase tuning, enables a crystal engraved with a single grating to be used instead of a parallelepipedal crystal engraved with a plurality of gratings. Another particularly advantageous aspect of the invention is that, in the case of said crystal with

the property of phase tuning by double refraction, tunability may be obtained on a larger spectral domain on the one hand, and the beam attenuation and deformation phenomena which may be observed under oblique incidence may be limited on the other hand, and in the case of said crystal with the property of quasi phase tuning, a more continuous tunability may be obtained.

The crystal of the device according to the invention may have a network of monocrystalline domains selected from a network of plane monocrystalline domains, a network of curved (circular, elliptical) monocrystalline domains. Parameters of such networks are further described in the examples which follow.

This crystalline network, whether plane or curved, may also have periodically alternating domains, notably for a quasi phase tuning operation: the network may then be made up of domains for which the effective coefficient's sign alternates periodically (a + domain, a - domain, ...), or for which the value of the refractive index is periodically modulated (an index domain n_1, an index domain $n_2 \neq n_2$, ...). In the case of a plane network, the volume delimited by this alternating network may also be advantageously surrounded by a monocrystalline crown c for which the effective coefficient's sign does not alternate. This may now be a periodically alternating network, optionally surrounded by a non-alternating monocrystalline crown c. A configuration of an

alternating plane network surrounded by a non-alternating crown c is illustrated in the following Example 11 and in Fig. 10. Alternatively, this may be a curved, circular or elliptical periodically alternating network. Such a curved alternating network is notably contemplated in order to prevent or limit losses by refraction or diffusion at the plane interfaces of the + zones and the - zones under oblique incidence. These configurations are illustrated and their parameters, notably for calculating the periodicity, are described in detail in the following figures and examples.

According to an embodiment of the invention, said incident optical radiation(s) comprise (each) one, two, three or four equal or different frequencies, with colinear or non-colinear wave vectors, and under normal or quasi normal incidence (deviation by an angle $i(\alpha)$ with respect to the normal, such as defined in the following Example 12 and in Fig. 11) on one of the surface(s) of said crystal defining a cylindrical volume of revolution. Such radiations may notably be generated by a source of electromagnetic radiations associated with said device.

Preferably, said incident optical radiation(s) are laser radiation(s), such as radiations emitted by a gas laser (helium-neon, ionized argon, nitrogen and carbon dioxide lasers, excimers, a solid laser (ruby, neodymium ions), a liquid dye laser, a semiconductor laser (gallium arsenide), a free electron laser. Notably, they may be laser radiation(s) selected from

laser radiation including one or more set frequencies and laser radiation including one or more tunable frequencies.

The device according to the invention is notably suitable for producing three-wave or four-wave interaction(s). Said crystal advantageously has a non-centrosymmetric structure so that said device provides three-wave interaction(s).

According to another embodiment of the invention, said (or at least one of said) incident optical radiation(s) comprise two frequencies for three-wave interaction or three frequencies for four-wave interactions, and said (or at least one of said) emerging optical radiation(s) comprise a frequency which corresponds to the sum of said two or if necessary of three frequencies comprised in said incident optical radiation(s).

According to another embodiment of the invention, said (or at least one of said) emerging optical radiation(s) comprise a frequency equal to a multiple and notably to the double or the triple of a frequency comprised in said (or at least one of said) incident optical radiation(s).

According to yet another embodiment of the invention, said (or at least one of said) incident optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, and said (or at least one of said) emerging optical radiation(s) comprise a

frequency which corresponds to a difference between said two, or if necessary three frequencies comprised in said incident optical radiation(s).

According to yet another embodiment of the invention, said (or at least one of said) emerging optical radiation(s) comprises two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, for which the sum is equal to a frequency comprised in said (or at least one of said) incident optical radiation(s).

Said (or at least one of said) interaction(s) may be an interaction with 4 colinear wave vectors or else an interaction with non-colinear wave vectors.

Said (or at least one of said) interaction(s) preferably are an interaction selected from an optical parametric amplification and a generation of second or third harmonics. The device according to the invention then operates as an optical parametric amplifier, or as a generator of second or third harmonics, respectively.

According to an advantageous embodiment of the invention, said crystal is placed inside a cavity for resonant interaction, and said optical system for confining and focussing said incident optical radiation(s) on the central portion(s) of said crystal on the one hand and for collimating and directing said emerging optical radiation(s) outside said cavity. This optical system in the device according to the invention is necessarily placed outside said cavity. The advantage of this configuration is such that losses by

refraction, diffraction, diffusion and absorption are less than those expected if there had been lenses between the crystal and the mirrors of the cavity. This resonant interaction may notably be a three- or four-wave interaction selected from an optical parametric oscillation, an optical parametric amplification and a generation of second or third harmonics. The configuration of said crystal inside said cavity is necessary for obtaining an optical parametric oscillation, but is optional for obtaining an optical parametric amplification or a generation of second or third harmonics. In the latter case, it actually aims at increasing the generation efficiencies. The device according to the invention may now operate as an optical parametric oscillator or as an optical parametric amplifier or as a generator of second or third harmonics, respectively.

According to an aspect of this advantageous embodiment, said cavity includes at least an input reflecting surface (receiving the incident radiation(s) and at least an output reflecting surface (receiving the emerging radiation(s)) facing each other providing resonance of at least one of the interacting waves. The geometry of said cavity is defined according to the specific refraction properties of "dioptres" (optical media with two interfaces) with a cylindrical volume of revolution of crystals with anisotropic optical properties, in particular with respect to the double refraction angle ρ, the angle between the Poynting

vector and the wave vector. The reflection coefficients of the input and output reflecting surfaces are such that the cavity may be resonant either with one, or with two, or with three or possibly with four interacting waves.

In particular, said input reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity. The distance between the input reflecting surface and said crystal is adapted in order to obtain the sought-after resonance(s).

If at least one of the resonant waves has a non-zero double refraction angle ρ, said output reflecting surface is placed at a distance d from said crystal and has a radius of curvature R, with a concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side of said crystal, so that the outgoing and returning beams of the resonant wave (or waves) coincide. The values of d and R advantageously obey equation $R = d - L$ with d greater than L for a concavity orientated on the side of said crystal or to equation $R = L - d$ with d less than L for a concavity orientated on the opposite side of said crystal, with L defined by $L = R_c\ (\cos(2\rho) + (\sin(2\rho)\ /\ \tan\ (\rho_e)) - 1)$, with R_c being the radius of

the cylindrical volume of revolution, ρ the angle of double refraction and with ρ_e defined by $\rho_e \cong \arcsin(n \sin(2\rho) - 2\rho)$, with n the index of refraction of said at least one wave for which resonance is sought after.

For resonant waves having a double refraction angle ρ equal to zero, said output reflecting surface may in particular be selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity. The distance between the output reflecting surface and said crystal is adapted in order to obtain the desired resonance(s).

The geometries of the cavity of the device according to the invention allow the laser beams to make round trips in the crystal without it being necessary to immerse this crystal in a refractive index adaptation medium or to place lenses between the crystal and the mirrors of the cavity. Thus, in the present invention, the level of losses by refraction, diffraction, diffusion and absorption is less than that obtained if there was an refractive index adaptation liquid around the crystal and/or the lenses between the crystal and the mirrors of the cavity. Because of a lower loss level, the oscillation threshold of the cavity is lower, i.e., the beams may be generated in

the non-linear crystal with lower intensity of the incident beam on the input mirror of the cavity.

According to another advantageous embodiment of the invention, said device further comprises means for thermostatically controlling said crystal. Such means notably may stabilize the efficiency of the interactions or extend the accessible spectral range. Said crystal is then advantageously held at a lower or higher temperature than the room temperature.

According to another advantageous embodiment of the invention, said device further comprises means for applying a static or low frequency electric field inside said crystal.

The geometry of the crystal, component of the device according to the invention, actually allows the laser radiation to be easily coupled with a static or low frequency electric field for modulating the efficiency of the interactions or for altering, and in particular for increasing, the accessible spectral range (via the electro-optical effect). Hence, said device may further comprise a pair of electrodes placed on the opposite faces of said crystal.

The device according to the invention has numerous applications, in particular civil or military applications for optical frequency-tunable radiations tunable. It thus advantageously forms a component selected from a spectroscope component, a remote detection component, a remote transmission system component or a remote guiding system component, a LIDAR

(Light Detection And Ranging) system component, an optronic counter-measure system component.

The present invention also aims at a method for generating one or more tunable optical radiation(s), at least in frequency, characterized in that it implements a device according to the invention.

An advantageous method according to the invention notably consists of:

i. generating one or more optical radiation(s) each comprising one or more spectral component(s),

ii. directing said optical radiation(s) as incident optical radiation(s) towards a so-called crystal under normal incidence on one surface or one of the surfaces of said crystal defining (independently of one another) a cylindrical volume of revolution, in order to generate, outside said crystal

- a frequency corresponding either to a multiple (double, triple) of a frequency of said incident radiation(s), or to the sum, or to a difference between the frequencies of said incident radiation(s), or still further

- several frequencies, the sum of which correspond to a frequency comprised in said incident optical radiation(s),

iii. if necessary, repeating items i and ii above after suitable rotation of said crystal around its axis of revolution or rotation of said incident optical radiations around said crystal in a plane orthogonal to the axis of revolution of said crystal.

According to a preferred embodiment of this advantageous method according to the invention, said (or at least one of said) incident optical radiation(s) is a laser radiation selected from laser radiation including one set frequency or set frequencies and laser radiation including one tunable frequency or tunable frequencies.

A particularly preferred embodiment of this advantageous method according to the invention further comprises the application of an electric field inside said crystal, in order to produce an electro-optical effect.

In the present application, reference is made to Figs. 1-13:

- Fig. 1 shows, on a cylindroid 2, the orthonormal axis system (x, y, z) and spherical coordinates (θ,φ) of an arbitrary direction,

- Fig. 2 illustrates different embodiments 1,2,3,4,5 of a crystal belonging to the device according to the invention, the hatched surfaces represent the surfaces of said crystals on which electrodes (electro-optical effect) may advantageously be placed, each reference number 1,2,3,4,5 designates a volume which may have a circular or elliptical section, each crystal is placed between two lenses L_1 and L_2,

- Fig. 3 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 having a completely cylindrical volume of revolution,

- Fig. 4 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 3 having a cylindrical volume of revolution in a truncated way on two opposite and symmetrical quadrants of angular deviation $\delta\alpha$ (truncated cylinder),

- Fig. 5 represents a device according to the invention comprising, placed between three couples of lenses (L_{1i}, L_{1e}), (L_{2i}, L_{2e}), and (L_{3i}, L_{3e}), a crystal with a non-linear optical property 1 having a completely cylindrical volume of revolution,

- Fig. 6 represents a device according to the invention comprising a crystal with a non-linear optical property 1 placed between two lenses L_1 and L_2, and placed inside a cavity having two input and output reflecting surfaces (or mirrors) providing resonance for the wave with Poynting vectors P_1,

- Figs. 7 and 8 represent a device according to the invention comprising a crystal with a non-linear optical property 1 placed between two lenses L_1 and L_2, and placed inside a cavity having two input and output reflecting surfaces (or mirrors) providing resonance for two waves with respective Poynting wave vectors P_1 and P_2; in Fig. 7, the input mirror is plane, the output mirror has a radius of curvature R with the concavity on the side of crystal L; in Fig. 8, the input mirror is plane, the output mirror has a radius of curvature R with the concavity on the opposite side to crystal 1,

- Fig. 9 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 having a completely cylindrical volume of revolution and having along the direction of propagation of a sought-after interaction, an effective non-linear coefficient with alternate signs (+, -) according to the periodicity vector V and with a period Δ (periodic plane network for quasi phase tuning),

- Fig. 10 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 with a circular section adapted to the quasi phase tuning and comprising two concentric zones: a central zone with a periodic plane network (modulation (+, -) of the sign of the effective non-linear coefficient) and a monocrystalline crown zone c for which the effective non-linear coefficient does not alternate (+ or -),

- Fig. 11 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 with an elliptical section adapted to quasi phase tuning, and comprising an elliptical network for which the effective non-linear coefficient has an alternating sign (+, -) according to a periodicity Δ which may vary from Δ_{min} to Δ_{max} over two angular sections of aperture β,

- Fig. 12 represents a device according to the invention, placed between two lenses L_1 and L_2, a

crystal with a non-linear optical property 5, with an elliptical section adapted to quasi phase tuning, corresponding to a portion of an ellipse of aperture β, between 0° and 180° where a network is inscribed for which the period may vary from Δ_{min} to Δ_{max},

- Fig. 13 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 with a periodic circular network for which the refractive indexes n+ or n- vary according to the direction of propagation in a given plane.

EXAMPLES

Example 1. <u>Calculation of the phase tuning directions through double refraction</u>

Phase tuning through double refraction provides optimization of the efficiency of optical parametric interactions. In the case of crystals where the angular frequency ($\omega = 2\pi v$ where v is the frequency) dispersion of refractive indexes (n) is said to be "normal", i.e., $n(\omega_i) < n(\omega_j)$ when $\omega_i < \omega_j$, and for interactions where the wave vectors of coupled waves are colinear, the different possible phase tuning relationships are the following:

- for interactions with 3 waves of angular frequencies ω_1, ω_2 and ω_3 (with $\omega_1 + \omega_2 = \omega_3$)

$$\omega_1 n^-(\omega_1,\theta,\phi) + \omega_2 n^-(\omega_2,\theta,\phi) = \omega_3 n^-(\omega_3,\theta,\phi) \quad (1a)$$

$$\omega_1 n^+(\omega_1,\theta,\phi) + \omega_2 n^-(\omega_2,\theta,\phi) = \omega_3 n^-(\omega_3,\theta,\phi) \quad (1b)$$

$$\omega_1\, n^-(\omega_1,\theta,\phi) + \omega_2\, n^+(\omega_2,\theta,\phi) = \omega_3\, n^-(\omega_3,\theta,\phi) \quad (1c)$$

- for interaction with four waves of angular frequencies ω_1, ω_2, ω_3 and ω_4 (with $\omega_1 + \omega_2 + \omega_3 = \omega_4$)

$$\omega_1\, n^+(\omega_1,\theta,\phi) + \omega_2\, n^-(\omega_2,\theta,\phi) + \omega_3\, n^-(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^-(\omega_1,\theta,\phi) + \omega_2\, n^-(\omega_2,\theta,\phi) + \omega_3\, n^+(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^-(\omega_1,\theta,\phi) + \omega_2\, n^+(\omega_2,\theta,\phi) + \omega_3\, n^-(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^-(\omega_1,\theta,\phi) + \omega_2\, n^-(\omega_2,\theta,\phi) + \omega_3\, n^+(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^+(\omega_1,\theta,\phi) + \omega_2\, n^+(\omega_2,\theta,\phi) + \omega_3\, n^-(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^+(\omega_1,\theta,\phi) + \omega_2\, n^-(\omega_2,\theta,\phi) + \omega_3\, n^+(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

$$\omega_1\, n^-(\omega_1,\theta,\phi) + \omega_2\, n^+(\omega_2,\theta,\phi) + \omega_3\, n^+(\omega_3,\theta,\phi) = \omega_4\, n^-(\omega_4,\theta,\phi)$$

For interactions where the wave vectors of coupled waves are non-colinear, the combinations of refractive indexes n^- and n^+ are identical to the colinear cases, but with different coefficients: for interactions with 3 waves, ω_1 is replaced with $\omega_1 \cos[\alpha_{13}(\theta,\phi)]$ and ω_2 is replaced with $\omega_2 \cos[\alpha_{23}(\theta,\phi)]$; for interaction with 4 waves, ω_1 is replaced with $\omega_1 \cos[\alpha_{14}(\theta,\phi)]$, ω_2 is replaced with $\omega_2 \cos[\alpha_{24}(\theta,\phi)]$ and ω_3 is replaced with $\omega_3 \cos[\alpha_{34}(\theta,\phi)]$, with ω_{ij} the angle between the wave vectors at ω_i and ω_j ($i = 1$ or 2 if $j = 3$; $i = 1$ or 2 or 3 if $j = 4$). For example, in the case of 3-wave interactions, the projection of the vector relationships for phase tuning on the direction of the wave vector 3 transforms relationship (1a) into:

$$\omega_1\cos[\alpha_{13}(\theta,\phi)]\,n^+(\omega_1,\theta,\phi) + \omega_2\cos[\alpha_{23}(\theta,\phi)]\,n^+(\omega_3,\theta,\phi) = \omega_3 n^-(\omega_3,\theta,\phi) \quad (3)$$

where $(\alpha_{13})(\alpha_{23})$ are the angles between the wave vectors

at ω_1 and at ω_3 (ω_2 and ω_3), respectively. (θ,ϕ) are the spherical coordinates of the phase tuning direction as seen in the optical referential (x, y, z) which is bound to the non-linear crystal. Fig. 1 illustrates the configuration of axes (x, y, z) and the spherical coordinates (θ,ϕ) of a arbitrary direction. Section (x,y) may be circular or elliptical. Spherical coordinates (θ,ϕ) are related to Cartesian coordinates (U_x, U_y, U_z) by:

$$U_x = \cos\phi \sin\theta;\ U_y = \sin\phi \sin\theta;\ U_z = \cos\theta$$

$n^+(\omega,\theta,\phi)$ and $n^-(\omega,\theta,\phi)$ are the solutions of Fresnel's equation and are given by:

$$n^{\pm} = \left(\frac{2}{B \mp \left(B^2 - 4C\right)^{1/2}} \right)$$

$$B = -u_x^2(b+c) - u_y^2(a+c) - u_z^2(a+b) \qquad C = u_x^2 bc + u_y^2 ac + u_z^2 ab$$

$$a = -n_x^{-2}(\omega) \qquad b = n_y^{-2}(\omega) \qquad c = n_z^{-2}(\omega)$$

the surface $n'(\theta,\phi)$ is called the index surface. $n_x(\omega)$, $n_y(\omega)$, $n_z(\omega)$ are the main indexes of refraction at angular frequency ω. The main indexes of refraction at the angular frequencies of the interacting waves are the parameters necessary for resolving the phase tuning equations (1), (2) and (3); they are given by Sellmeier's equations with 4, 5 or 6 coefficients. Sellmeier's equations for the crystals considered hereafter in Examples 2, 3, 5 and 6 are given at room temperature here. They are determined from several measurements of refractive indexes at different

wavelengths λ (λ is expressed in μm in equations (5) to (11) below).

KTiOPO₄:

$$n_x^2(\lambda) = 2.1239 + \frac{0.142744\lambda^2}{\lambda^2 - 18.477} + \frac{0.87370\lambda^2}{\lambda^2 - 0.045906}$$

$$n_y^2(\lambda) = 2.0649 + \frac{0.15529\lambda^2}{\lambda^2 - 19.373} + \frac{0.95463\lambda^2}{\lambda^2 - 0.045505}$$

$$n_z^2(\lambda) = 1.6539 + \frac{0.34767\lambda^2}{\lambda^2 - 29.378} + \frac{1.6482\lambda^2}{\lambda^2 - 0.038825}$$

RbTiOPO₄:

$$n_x^2(\lambda) = 2.15559 + \frac{0.93307\lambda^2}{\lambda^2 - 0.044075} - 0.01452\lambda^2$$

$$n_y^2(\lambda) = 2.38494 + \frac{0.73603\lambda^2}{\lambda^2 - 0.057078} - 0.01583\lambda^2$$

$$n_z^2(\lambda) = 2.27723 + \frac{1.11030\lambda^2}{\lambda^2 - 0.055009} - 0.01995\lambda^2$$

RbTiOAsO₄:

$$n_x^2(\lambda) = 2.04207 + \frac{1.17785\lambda^2}{\lambda^2 - 0.040630} - 0.01035\lambda^2$$

$$n_y^2(\lambda) = 2.14941 + \frac{1.09267\lambda^2}{\lambda^2 - 0.046062} - 0.01067\lambda^2.$$

$$n_z^2(\lambda) = 2.18962 + \frac{1.30103\lambda^2}{\lambda^2 - 0.052025} - 0.01390\lambda^2$$

CsTiOAsO₄:

$$n_x^2(\lambda) = 2.34498 + \frac{1.04863\lambda^2}{\lambda^2 - 0.048594} - 0.01483\lambda^2$$

$$n_y^2(\lambda) = 2.74440 + \frac{0.70733\lambda^2}{\lambda^2 - 0.067772} - 0.01526\lambda^2$$

$$n_z^2(\lambda) = 2.53666 + \frac{1.10600\lambda^2}{\lambda^2 - 0.062440} - 0.01711\lambda^2$$

KTiOAsO₄:

$$n_x^2(\lambda) = 2.8049 + \frac{0.35190\lambda^2}{\lambda^2 - 0.098915} - \frac{0.27186\lambda^2}{\lambda^2 + 15.798}$$

$$n_y^2(\lambda) = 2.8077 + \frac{0.37614\lambda^2}{\lambda^2 - 0.093917} - \frac{0.22531\lambda^2}{\lambda^2 + 8.6981}$$

$$n_z^2(\lambda) = 3.8510 + \frac{0.81874\lambda^2}{\lambda^2 - 86.976} - \frac{0.44017\lambda^2}{\lambda^2 + 28229}$$

$LiNbO_3$:

$$n_o^2(\lambda) = 4.9048 + \frac{0.11775\lambda^2}{\lambda^2 - 0.047533} - 0.027153\lambda^2$$

$$n_e^2(\lambda) = 4.5820 + \frac{0.09921\lambda^2}{\lambda^2 - 0.044479} - 0.021940\lambda^2$$

$LiTaO_3$:

$$n_e^2(\lambda) = 4.5284 + \frac{0.0095478\lambda^2}{\lambda^2 - 0.060858} + \frac{0.077690\lambda^2}{\lambda^2 - 0.033782} - 0.023670\lambda^2$$

not measured (not requested)

$n_0^2(\lambda)$

For a given interaction, characterized by the angular frequencies of the interacting waves, there is a set of phase tuning directions, i.e., couples (θ,ϕ) which are solutions of the same phase tuning equation. Furthermore, these different phase tuning directions are not equivalent from the point of view of the interaction efficiency. Moreover, the phase tuning directions are different from one interaction to another. Thus, an arbitrary orientation plane contains a set of phase tuning directions. The interesting planes for the present invention are in particular certain planes of symmetry of the index surface. These planes are orthogonal to the axis of revolution of the cylinder.

For a uniaxial crystal, defined by $n_x = n_y \neq n_z$, the useful planes are the orthogonal planes to plane (xy), i.e., at constant ϕ angle: each of these planes contains phase tuning directions which are symmetrical

two by two with respect to the z axis, and each of these couples corresponds to a particular interaction. The different planes, corresponding to different ϕ values, have the same phase tuning spectral range but are distinguished by the interaction efficiency.

For a biaxial crystal, defined by $n_x \neq n_y \neq n_z$, the three main planes (xy), (xz) and (yz) are of interest. Plane (xy) contains the phase tuning directions which are symmetrical two by two with respect to x and y, each of the couples corresponding to a specific interaction. Planes (xz) and (yz) have the same symmetry than the planes of an uniaxial crystal containing the z axis as described earlier. The three planes (xy), (xz) and (yz) of the biaxial crystal do not have the same phase tuning spectral ranges.

Example 2: <u>Forms of the non-linear crystal</u>

Fig. 2 illustrates various embodiments of a crystal belonging to the device according to the invention, the hatched surfaces represent surfaces of said crystal on which electrodes may be advantageously placed (for the electro-optical effect). Reference numbers 1 and 2, each refer to a crystal with a non-linear optical property having a completely cylindrical volume of revolution (reference 1: cylinder; reference 2: cylindroid), reference 3 and 4 refer to a crystal with a non-linear optical property having a cylindrical volume of revolution in a truncated way on opposite and symmetrical quadrants with respect to its axis of revolution, (reference 3: truncated cylinder; reference 4: truncated cylindroid), reference 5 refers to a crystal with a non-linear optical property corresponding to a portion of said volumes 1 or 2 along

planes containing the axis of revolution (reference 5: partial cylinder or cylindroid). Said volumes of cylinder 1 and cylindroid 2 may have a circular section or an elliptical section; such a section may notably be considered along an orthogonal plane to the axis of revolution. Electrodes may be advantageously placed on both sides of a crystal according to different configurations, they are illustrated in Fig. 2 by a pair of hatched surfaces. For the crystals of volume 3 or 4, two of these configurations are illustrated: a pair of horizontally orientated electrodes or a pair of vertically orientated electrodes.

Example 3: <u>Configuration and geometry of the components for focussing and collecting radiation</u>

The incident radiation(s) are focussed on the crystal by one or more lenses or by a mirror with a concavity orientated on the side of said crystal or by a mirror with a concavity orientated on the opposite side of said crystal. The emerging radiation(s) are collected by one or more lenses or by a mirror with a concavity orientated on the side of said crystal or by a mirror with a concavity orientated on the opposite side of said crystal, regardless of the focussing device.

The focal distance of the focussing device is such that the incident radiation(s) have a small dimension with respect to the radius of the crystal in order to limit optical aberrations and for increasing the interaction efficiency, and such that this dimension is sufficient for preventing any damage to the crystal by a too strong incident intensity. The focal distance of the device collecting the emerging radiation is such

that the divergence of this emerging radiation is minimum.

These focal distances are between about 50 and 500 mm. Figs. 3 and 4 illustrate the realization of a device according to the invention where only one incident radiation is focussed on the crystal. Fig. 3 represents a device according to the invention comprising a crystal with a non-linear optical property 1, having a completely cylindrical volume of revolution and a focal distance f. This crystal is capable of rotating by an angle α which may range from 0° to 360°. It is placed between two lenses L_1 and L_2 with focal distances f_1 and f_2, respectively.

Fig. 4 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 3, having a cylindrical volume of revolution in a truncated way on two opposite and symmetrical quadrants of angular deviation $\Delta\alpha$.

Fig. 5 illustrates the realization of a device according to the invention where three incident radiations are focussed on the crystal. The crystal with a non-linear optical property 1 has a completely cylindrical volume of revolution. It is placed between 6 convergent lenses; L_{1i}, L_{2i}, L_{3i} are the lenses used for focussing the different incident radiations; L_{1e}, L_{2e}, L_{3e} are the lenses used for collecting the different emerging radiations.

Example 4: <u>General calculation of a cavity</u>

In the case of resonant interactions, the crystal is placed inside a cavity. The geometry of the cavity, the radius of curvature of mirrors R and distances

separating the mirrors of crystal d, is defined according to the specific refraction properties of cylindrical dioptres of crystal with anisotropic optical properties, in particular as regards the angle ρ for double refraction, the angle between the Poynting vector (the beam) and the wave vector (the wave). The reflection coefficients of the input and output mirrors are such that the cavity may be resonant either with one or two or three or four interacting wave(s).

The mirror receiving the incident radiation, called the input mirror, may be plane. In order to optimize the oscillation threshold and the stability of the cavity, the input mirror may have a suitable radius of curvature (for example a spherical, cylindrical or parabolic mirror), with the concavity on the side of the non-linear crystal or on the opposite side of the non-linear crystal.

If the resonant waves have zero double refraction angle, the mirror receiving the emerging radiation, called the output mirror, may be plane as is shown in Fig. 6.

Fig. 6, illustrates a device according to the invention comprising a crystal with a non-linear optical property 1 placed between two lenses L_1 and L_2, and placed inside a cavity having two input and output reflecting surfaces (or mirrors) providing resonance of the wave, for which the Poynting vector is P_1. The lenses are placed on the outside of the cavity. The diagram of Fig. 6 only shows two types of beams: the resonant beam represented by the Poynting vector P_1, for which angle ρ is zero, and the non resonant beam represented by the Poynting vector P_2, with non-zero ρ. The wave vector associated with P_1 and P_2, are colinear

with P_1. Only one round trip in the cavity is illustrated: + direction for the outward travel, - direction for the return. In order to optimize the oscillation threshold and the stability of the cavity, the output mirror may have a suitable radius of curvature (for example a spherical, cylindrical or parabolic mirror) with the concavity on the side of the non-linear crystal or with the concavity orientated on the opposite side of the non-linear crystal.

If at least one of the resonant waves at a non-zero double refraction angle ρ, the output mirror is spherical or cylindrical with radius R. The selection of the orientation, with respect to the crystal and the magnitude of the radius of curvature R as well as the distance d separating the crystal from the output mirror depends on the geometrical parameter L in such a way that the outgoing and returning beams of the resonant wave coincide as shown in Figs. 7 and 8.

Figs. 7 and 8 actually represent a device according to the invention comprising a crystal with a non-linear optical property 1 placed between two lenses L_1 and L_2, and placed inside a cavity having two input and output reflecting surfaces (or mirror) providing resonance of two waves with their wave vectors colinear with P_1, represented by their respective Poynting vectors P_1 and P_2, with P_1 being resonant or non resonant, and such that the corresponding double refraction angle ρ is zero, with P_1 resonant such that the corresponding double refraction angle ρ is non-zero. The lenses are placed outside the cavity. In Fig. 7, the input mirror is plane, the output mirror has a radius of curvature R with the concavity on the side of the crystal 1; in Fig. 8, the input mirror is

plane, the output mirror has a radius of curvature R with the concavity on the opposite side to crystal 1.

Parameter L is defined by:

(12) $$L = R_c\left[\cos 2\rho + \frac{\sin 2\rho}{\tan \rho_e} - 1\right]$$

avec $\rho = \arcsin(n \sin 2\rho) - 2\rho$ (13)

n is the index of refraction of the resonant wave for which angle ρ is non-zero.

Figs. 7 and 8 only show two types of beams: beam P_1, either resonant or not, the polarization of which is such that angle ρ is zero and beam P_2, resonant, with non-zero ρ. The wave vectors associated with P_1 and P_2, are colinear with P_1. Only one round trip is illustrated in the cavity: + direction for the outward travel, - direction for the return.

Fig. 7 concerns an output mirror, the concavity of which is orientated on the side of the crystal. For a given d value, the radius of curvature R is defined by:

$$R = d - L \quad \text{with } d > L \qquad (14)$$

L is given by relationships (12) and (13).

Fig. 8 concerns an output mirror with the concavity orientated on the opposite side of the crystal.

For a given d value, the radius of curvature R is defined by:

$$R = L - d \quad \text{with } d < L \qquad (15)$$

L is given by relationships (12) and (13).

The ratio L/R_0 is constant to within 10^{-3} for values of ρ between 0° and 2°, which are the typical values for most of the crystals which may be used for parametric interactions. The variation of L/R_0 is of the order of 10^{-2} when ρ varies from 2° to 3°. Thus,

for these cases with a large double refraction angle, the paths of the outgoing and returning beams coincide if the output mirror has a suitable curvature, is non-spherical, and non-cylindrical, this takes into account the values of L and ρ corresponding to the spectral range used; such a mirror may be used in order to optimize the stability and the oscillation threshold of the cavity.

For a mirror with a given radius of curvature R, the configuration according to Fig. 8 may only be contemplated if the radius of the cylinder is greater than a minimum value.

$$R_c > \frac{R}{\cos 2\rho + \dfrac{\sin 2\rho}{\tan \rho_e} - 1} \qquad (16)$$

Such that d (= L - R) is positive.

When both configurations of Figs. 7 and 8 may be contemplated, the case of Fig. 8 leads to a shorter total length of the cavity ($2R_c+L$ for two input and output mirrors with the same radius of curvature) than for the configuration of Fig. 7 ($2R_c+L+2R$). For radiations with short time pulses, a cavity with a small length provides a more efficient conversion.

Example 5: <u>Phase tuned optical parametric oscillator (OPO), by double refraction.</u>

In the case of an interaction with three waves of respective angular frequencies ω_1, ω_2 and ω_3 with $\omega_3 = \omega_1 + \omega_2$, the incident laser radiation, called the "pump" has a wavelength of $\lambda_p = 2\pi c/\omega_3$, where in c is the speed of light *in vacuo*, ω_3 is the largest of the three angular frequencies of the interacting waves. The

two angular frequencies of the waves emitted in the OPO are called "signal" and "complementary", with respective wavelengths $\lambda_s = 2\pi c/\omega_2$, and $\lambda_i = 2\pi c/\omega_1$, such that $\omega_1 + \omega_2 = \omega_3$, and $\omega_1 < \omega_2$. Table 1 below gives for $\lambda_p = 0.532\ \mu m$ and $\lambda_p = 1.064\ \mu m$, the range of wavelengths of the signal $\Delta\lambda_s$ and of the complementary $\Delta\lambda_i$, which may be generated with phase tuning by double refraction for a total rotation of $\Delta\alpha$, from the phase tuning minimum angle α_{min} for cylinders of several crystals with non-linear optical properties: $KTiOPO_4$ (KTP), $CsTiOAsO_4$ (CTA), $RbTiOPO_4$ (RTP), $RbTiOAsO_4$ (RTA), $KTiOAsO_4$ (KTA), $LiNbO_3$. The axis of revolution, X, Y or Z, is an axis of the optical reference system, as defined in Example 1. For a rotation around the Z axis, α is the angle ϕ in spherical coordinates; for a rotation around axes Y or X, α corresponds to the angle θ in spherical coordinates. $\Delta\alpha$ and α_{min} are calculated from the phase tuning relationships and Sellmeier's equations from Example 1.

Each crystal, machined into a complete cylinder 1, into a truncated cylinder 3 or into a partial cylinder 5 (cf. Fig. 2) is placed inside a resonant cavity such as defined above in Example 1 (lenses outside the cavity).

Table 1

Crystal	λp (μm)	Δλs (μm)	Δλi (μm)	Axis of rotation of the cylinder	Angle of rotation Δα (°)	αmin (°)
$KTiOPO_4$	0.532(-)	0.62-1.04(+)	1.09-3.5(-)	Y	46	44
$KTiOPO_4$	1.064(-)	1.53-2.12(+)	2.13-3.5(-)	Y	7	45.5
$CsTiOAsO_4$	0.532(-)	0.59-0.74(+)	1.9-5.2(-)	Y	39	51
$CsTiOAsO_4$	1.064(-)	1.33-2.12(+)	2.13-5.2(-)	Y	15	61.5
$RbTiOAsO_4$	0.532(-)	0.59-0.855(+)	1.41-5.2(-)	Y	49	41
$RbTiOAsO_4$	1.064(-)	1.33-2.12(+)	2.13-5.2(-)	Y	9.3	44.6
$RbTiOPO_4$	0.532(-)	0.62-0.95(+)	1.21-3.5(-)	Y	45	45
$RbTiOPO_4$	1.064(-)	1.52-2.12(+)	2.13-3.5(-)	Y	8.3	49.5
$LiNbO_3$	0.532(-)	0.59-0.70(+)	2.2-5.2(-)	Y(or X)	40	50
$LiNbO_3$	1.064(-)	1.33-2.12(+)	2.13-5.2(-)	Y(or X)	21	57
$KTiOPO_4$	0.532(-)	0.95-1.06(-)	1.06-1.2(+)	Z	67	23
$KTiOPO_4$	1.064(-)	1.52-1.58(-)	3.26-3.5(+)	Z	90	0
$CsTiOAsO_4$	0.532(-)	0.74-0.84(+)	1.45-1.9(-)	Z	90	0
$CsTiOAsO_4$	1.064(-)	1.7-2.0(-)	2.27-2.8(+)	Z	90	0
$RbTiOAsO_4$	0.532(-)	0.855-0.955(+)	1.2-1.41(-)	Z	90	0
$RbTiOAsO_4$	1.064(-)	1.54-1.61(-)	3.15-3.4(+)	Z	90	0
$RbTiOPO_4$	0.532(-)	0.95-1.06(+)	1.06-1.21(-)	Z	60	0
$RbTiOPO_4$	1.064(-)	1.61-1.69(-)	2.85-3.15(+)	Z	90	0
$KTiOAsO_4$	0.532(-)	0.96-1.04(+)	1.08-1.19(-)	Z	90	0
$KTiOAsO_4$	1.064(-)	1.55-1.60(-)	3.15-3.38(+)	Z	90	0

(+) and (-) refer to indexes of refraction n^+ and n^- defined in Example 1, which are involved in the phase tuning relationship.

The case of a tunable OPO in the range 1.4 μm - 5.2 μm for applications in infrared spectroscopy is detailed hereafter. The pump laser emits radiation at 0.532 μm (a YAG-Nd laser with doubled frequency). A crystal of $PbTiOAsO_2$ is machined into a partial

cylinder 3 (cf. Fig. 2) with optically polished cylindrical faces. The axis of revolution is borne by axis Y (identical with the crystallographic axis b). The extreme directions of the cylindrical portion are axis X (identical to the crystallographic axis a) on the one hand and the direction ($\phi = 0°$, $\theta = 41°$) on the other hand. Rotation of the crystal is performed around axis Y between these two extreme directions by means of a motor-driven micrometric rotary device secured to the crystal.

The radius of the cylindrical portion is $R_0 = 10$mm. The crystal is placed between two cylindrical mirrors with radius of curvature R = 10mm. A mirror is totally reflecting for wavelengths from 0.59 μm to 0.86 μm, the second mirror has a large reflection coefficient (**R** = 90%) for the same wavelength. The mirrors are positioned according to the layout described in Fig. 8. The total length of the cavity is 32.5mm with L = 12.5mm and d = 2.5mm. If the mirrors are positioned according to the layout described in Fig. 7, the total length of the cavity is 52.5mm with L = 12.5mm and d = 22.5mm.

Example 6: <u>Phase tuned second harmonic generator (GSH) by double refraction</u>

In the case of an interaction with 3 waves of respective angular frequencies ω_1, ω_2 and ω_3, with $\omega_3 = \omega_1 + \omega_2$, the incident so-called "pump" radiation has a wavelength of $\lambda_p = 2\pi c/\omega$, with $\omega = \omega_1 = \omega_2$. The

angular frequency of the emitted waves in the GSH is the second harmonic of the pump wave, i.e., a wavelength $\lambda_h = 2\pi c/2\omega$, with $2\omega = \omega_3$. Table 2 gives for a total rotation of $\delta\alpha = 90°$, the range of pump wavelengths $\Delta\lambda_p$ for which phase tuning by double refraction of the GSH is possible in cylinders of several crystals with non-linear optical properties: $KTiOPO_4$ (KTP), $RbTiOAsO_4$ (RTA), $CsTiOAsO_4$ (CTA), $RbTiOPO_4$ (RTP), and $KTiOAsO_4$ (KTA). Rotation is performed around the Z axis of these crystals (the crystallographic axis c which is the binary axis), from one of the X or Y axes. The calculations are carried out from the phase tuning relationships 1b (equivalent to 1c in the case of GSH) of Example 1. Each crystal, machined into a complete cylinder 1, into a truncated cylinder 3 or into a partial cylinder 5 (cf. Fig. 2) is placed outside or inside a cavity as defined in Example 4 above.

Table 2

Crystal	$\Delta\lambda_I$ (μm)
$KTiOPO_4$	0.99-1.08
$RbTiOAsO_4$	1.14-1.25
$CsTiOAsO_4$	1.27-1.55
$RbTiOPO_4$	1.04-1.15
$KTiOAsO_4$	1.08-1.15

The case of a cylinder of $CsTiOAsO_4$ is detailed hereafter. It is very difficult to obtain spectrally

refined radiation between 0.5 μm and 0.7 μm with conventional OPOs. For applications in spectroscopy in this wavelength domain, doubling the frequency of the radiation emitted between 1 μm and 1.4 μm by an OPO is contemplated. A crystal of $CsTiOAsO_4$ machined into a partial cylinder 3 (cf. Fig. 2) with the binary Z axis (crystallographic c axis) as axis of revolution is fixed on a motor-driven micrometric rotary support. The cylindrical face of the crystal is optically polished. The incident radiation, emitted between 1.27 μm and 1.55 μm by a OPO, is focussed by a convergent lens with focal length 100mm (placed outside the cavity) into the crystal of radius R = 5mm. The emerging radiation is collected with a second lens with identical focal length, also placed outside the cavity as shown in Example 4. With a complete rotation of 90° between the X and Y axes, harmonic radiation between 0.63 μm and 0.77 μm may be generated efficiently because of the double refraction angles which are always small for the considered phase tuning directions.

Example 7: Quasi-phase tuning: calculation of the coherence length

The advantage of the quasi-phase tuning is notably that it may be achieved for combinations of any n^+ or n^- solutions with the angular frequencies of the interacting waves. $n^\pm$ are given by the relationships (4) from Example 1.

The coherence length L_c of the parametric

interactions in the crystals with the periodically alternating structure from Examples 9 and 10 is given by the following relationship in the case of an interaction with three waves of angular frequencies ω_1, ω_2 and ω_3:

$$L_c = \frac{\pi c}{\omega_3 n(\omega_3) - [\omega_1 n(\omega_1) + \omega_2 n(\omega_2)]} \tag{17}$$

n is calculated from the Sellmeier equations given in Example 1. In a structure with period 2Lc, the frequency conversion interaction is quasi phase tuned when the indexes of refraction of the interacting waves $n(\omega_1)$, $(n\omega_2)$ and (ω_3) satisfy the relationship (17).

Example 8: <u>Calculation of the variation of the coherence length as a function of the angle of rotation of the crystal in the case of a periodic plane network</u>.

Fig. 9 represents a device according to the invention comprising, placed between two lenses L_1 and L_2, a crystal with a non-linear optical property 1 having a completely cylindrical volume of revolution, capable of rotating by an angle which may range from 0 to 360° and having an effective non-linear coefficient with alternating signs (+, -) according to the periodicity vector V. p is the width of each monocrystalline domain of the network along V. As the laser radiation is considered fixed, the coherence length along the propagation direction is a function of the angle of rotation α. It is given by the following

relationship:

$$L_c(\alpha) = \frac{P}{\cos\alpha} \tag{18}$$

The periodicity of the network therefore varies in function of α according to the following relationship:

$$\Delta\alpha = 2 \times L_c(\alpha) \quad (19)$$

Thus, to each angle α corresponds a particular parametric interaction (ω_1, ω_2, ω_3) such as $L_c(\alpha)$ is equal to an odd multiple of the coherence length of this interaction. The tunability of the parametric device is thus achieved. The symmetry of the periodic structure is such that a total rotation of $\delta\alpha = 90°$ enables access to all the periodicities: $\Delta(\alpha=0°) = 2p$ to $\Delta(\alpha=90°) \rightarrow \infty$. Thus, a cylindrical volume of revolution described in a truncated way 3 (cf. Fig. 2) or in a partial way (portion 5 of the cylinder) may also be used in this case. The same argument is also valid for a volume of a cylindroid 2 with respect to a volume of a truncated cylindroid 4, and a volume 5 of a partial cylindroid. This also applies whether the (xy) section is circular or elliptical.

Example 9: <u>Quasi phase tuned optical parametric oscillator (OPO)</u>

In the case of an interaction with 3 waves of respective angular frequencies ω_1, ω_2 and ω_3 with $\omega_3 = \omega_1 + \omega_2$, the so-called "pump" incident laser radiation has a wavelength of $\lambda_p = 2\pi c/\omega_3$, wherein c is the speed of

light *in vacuo* and ω_3 is the largest of the three angular frequencies of the interacting waves. The two angular frequencies of the waves emitted in the OPO are called "signal" and "complementary", with respective wavelength $\lambda_3 = 2\pi c/\omega_3$, $\lambda_1 = 2\pi c/\omega_1$, such that $\omega_1 + \omega_2 = \omega_3$ with $\omega_1 < \omega_2$. Table 3 below gives for $\lambda_p = 0.532$ μm and $\lambda_1 = 1.064$ μm, the range of wavelengths for the signal and the complementary, respectively $\Delta\lambda_n$ and $\Delta\lambda_1$, which may be generated, quasi phase tuned for a total rotation of $\delta\alpha$ of the cylinder of several crystals with non-linear optical properties, $KTiOPO_4$, $CsTiOASO_4$, $RbTiOAsO_4$, $LiNbO_3$, $LiTaO_3$, $RbTiOPO_4$, and $KTiOAsO_4$, wherein the effective coefficient's sign periodically alternates over a coherence length L_c (cf. Fig. 9). The domains are inverted along the polar axis of the crystals which corresponds to the axis of revolution of the cylinder in accordance with what is described in Examples 7 and 8 above. The angle of rotation is located with respect to the periodicity vector of the network V. The relevant interaction relates to 3 polarized waves along the polar axis. The calculations are carried out from the relationship (17) of Example 7 and from Sellmeier's equations from Example 1.

Each crystal, machined into a complete cylinder 1 or into a truncated cylinder 3 or into a portion 5 of a cylinder (cf. Fig. 2), is placed in a resonant cavity such as defined earlier in Example 1. As the three interacting waves have a zero angle of refraction, the

input and output mirrors of the cavity may be plane.

Table 3

Material	λp (μm)	Δλs (μm)	Δλi (μm)	Pitch of the grating	Maximum angle of rotation δα (°)
$KTiOPO_4$	0.532	0.62-1.06	1.06-3.5	9.3	50
$KTiOPO_4$	1.064	1.52-2.12	2.13-3.5	36.5	23
$RbTiOAsO_4$	0.532	0.59-1.06	1.06-5.2	15.6	57
$RbTiOAsO_4$	1.064	1.33-2.12	2.13-5.2	34.8	34
$RbTiOPO_4$	0.532	0.62-1.06	1.06-3.5	8.8	49
$RbTiOPO_4$	1.064	1.52-2.12	2.13-3.5	32	25
$LiNbO_3$	0.532	0.59-1.06	1.06-5.2	6.8	56
$LiNbO_3$	1.064	1.33-2.12	2.13-5.2	25.7	36
$CsTiOAsO_4$	0.532	0.59-1.06	1.06-5.2	7.9	56
$CsTiOAsO_4$	1.064	1.33-2.12	2.13-5.2	30	38
$KTiOAsO_4$	0.532	0.59-1.06	1.06-5.2	8.5	55
$KTiOAsO_4$	1.064	1.33-2.12	2.13-5.2	29.5	36

The case of a tunable OPO between 3 μm and 5 μm for applications in optronic counter-measures is detailed hereafter. A structure of $LiNbO_3$ with alternating ferroelectic domains with periodicity 26.26 μm is machined into a truncated cylinder 3 (cf. Fig. 2), with optically polished cylindrical phases. The axis of rotation of the cylinder is the binary Z axis to which the ferroelectric domains are parallel. The periodicity vector V of the structure is perpendicular to this axis and is one of the extreme directions of the cylindrical portion. The other extreme direction of the cylindrical portion is located at 40.5° from the first. The structure is fixed on a motor-driven micrometric rotary support. The portion of

the cylinder has a radius R = 13mm and a thickness of 1mm. It is placed at the center of a cavity formed with two cylindrical mirrors with a radius of curvature R = 15mm, the concavity of which is orientated on the side of the crystal. The first mirror is totally reflecting for wavelengths between 1.35 μm and 1.65 μm, the second mirror has a reflection coefficient **R** = 90% for these same radiations. The total length of the cavity is 30mm. The pump laser radiation is emitted at 1.064 μm by a YAG:Nd laser. Two lenses with focal length f = 100mm are placed on both sides of the cavity in order to focus the incident radiation and to collect the emerging radiation.

Example 10: <u>Generator of a quasi-phase tuned second harmonic (GSH)</u>

In the case of an interaction with 3 waves of respective angular frequencies ω_1, ω_2 and ω_3 with $\omega_3 = \omega_1 + \omega_2$, the so-called "pump" incident radiation has a wavelength of $\lambda_p = 2\pi c/\omega$, with $\omega = \omega_1 = \omega_2$. The angular frequency of the waves emitted in the GSH is the second harmonic of the pump wave, i.e. a wavelength of $\lambda_G = 2\pi c/2\omega$, with $2\omega = \omega_3 + \omega_2$. Table 4 below gives the range of pump wavelengths $\Delta\lambda_F$ corresponding to a total rotation of $\delta\alpha$, for which quasi phase tuning of the GSH is possible in cylinders of several crystals with non-linear optical properties, $KTiOPO_4$, $RbTiOAsO_4$, $CsTiOASO_4$, $RbTiOPO_4$, $KTiOAsO_4$, $LiNbO_3$, and $LiTaO_3$, the

effective coefficient's sign periodically alternates over a coherence length L_c (cf. Fig. 9). The domains are inverted along the pole axis of the crystals which corresponds to the axis of revolution of the cylinder in accordance with what is described in Examples 7 and 8 above. The angle of rotation is located with respect to the periodicity vector V of the network. The relevant interaction relates to 3 polarized waves along the polar axis. The calculations are carried out from the relationship (17) of Example 7 and from the Sellmeier equations of Example 1.

Each crystal, machined into a complete cylinder 1 or into a truncated cylinder 3 or into a portion 5 of a cylinder (cf. Fig. 2), is placed either outside or inside a cavity such as defined above in Example 4. Fig. 6 illustrates such a device when the crystal is placed inside such a cavity. The focussing and collimating optical system (lenses) is then placed outside this cavity.

Table 4

Material	$\Delta\lambda_\varepsilon$ (μm)	Periodicity of the structure $V_\Delta=2L_c$ (μm)	Maximum angle of rotation $\delta\alpha$ (°)
$KTiOPO_4$	0.8-0.95	3.5	56
$RbTiOPO_4$	0.8-0.95	3.15	58
$RbTiOAsO_4$	0.8-0.95	3.0	58
$LiNbO_3$	0.75-0.95	2.0	64
$LiTaO_3$	0.64-0.95	1.3	76
$CsTiOAsO_4$	0.8-0.95	2.7	60
$KTiOAsO_4$	0.8-0.95	3.0	59

For example, a cylinder of $LiTaO_3$ with periodically alternating domains may be used for generating a tunable laser radiation of a short wavelength. The structure has a periodicity of 1.31 μm, a radius R = 8mm and a thickness of 0.5mm. The rotation of the structure is performed around the binary Z axis, on a micrometric rotary support. It enables the frequency of the incident laser radiation emitted by a titanium-sapphire laser or by a tunable OPO, to be doubled.

Example 11: Non linear crystal with crown for quasi-phase tuning

For certain materials, notably for those likely to exhibit brittleness at the interface between the + zone and the - zone, it is contemplated not to modulate the sign of the effective non-linear coefficient in the peripheral portion of the volume of the cylinder or cylindroid. This new configuration of the network is illustrated by Fig. 10.

The non-linear medium is then formed by two concentric zones: the central zone is modulated (+, -), whereas the peripheral zone (crown C) has a non alternating effective non-linear coefficient, i.e., either + or -. This crown may for example have a width ρ of 1 mm for an alternating central zone of diameter 25 mm. The advantage of this crown is to provide a good quality polished surface.

Example 12: <u>Calculation of the variation of the coherence length as a function of the angle of rotation in the case of a periodic elliptical network.</u>

An elliptical network instead of a plane network is also contemplated in the case when there are notably losses through refraction or diffusion at the plane interfaces of the + and - zones under oblique incidence.

Fig. 11 describes a device with an electrical curved network for quasi phase tuning according to the present invention. The cylindroid with axis of revolution O has an elliptical contour with major axis OA and minor axis OB. In two angular sectors facing each other with an aperture β which may be between 0° and 180°, identical networks are inserted, formed of elliptical crowns the axis of which coincide with OA and OB. These crowns are alternately + and - with a period given according to a radius of the ellipse located by angle α, $\Delta(\alpha) = 2L_c(\alpha)$; $\Delta(\alpha)$ varies in function of α from Δ_{min} to Δ_{max}, thus providing quasi phase tuning for frequency conversion interactions for which the coherence length $L_c(\alpha)$ is between $Lc_{min} = P_{min}$ and $Lc_{max} = P_{max}$. If α is located with respect to the major axis OA and for β = 90°, the coherence length in function of α is given by the following relationship:

$$L_c(\alpha) = \frac{(2D)}{\sqrt{\dfrac{\cos^2\alpha}{P^2_{max}} + \dfrac{\sin^2\alpha}{P^2_{min}}}}$$

Thus, the tunability of the device is achieved by rotation of the ellipse around O. When the radiations propagate along an α axis of the ellipse, they form an angle $i(\alpha)$ with the normal of the surface unit of the network, which is given by the following formula:

$$i(\alpha) = Arc\cos\left[\frac{P_{\max}^{2} + P_{\min}^{2}}{\sqrt{\dfrac{\cos^{2}\alpha}{P_{\max}^{4}} + \dfrac{\sin^{2}\alpha}{P_{\min}^{4}}}}\right] \qquad (21)$$

For example, let us consider a OPO according to the present invention with a structure of $LiNbO_3$ with alternating ferroelectric domains forming an elliptical network with P_{min} = 26 μm and P_{max} = 31.75 μm. The angle of incidence $i(\alpha)$ calculated from formula (21) is equal to 0° along the major axis (α = 0°) or along the minor axis (α = 90°) and it reaches a maximum value of the order of 11° around α = 45°. When this device is pumped at λ_p = 1.064 μm, the rotation of the ellipse by an angle α between 0° and 90° enables a radiation to be generated with λ_s between 1.33 μm and 2.12 μm and a radiation to be generated with λ_i between 2.13 μm and 5.2 μm. The larger $i(\alpha)$, the larger are the losses at the interface. Thus, the advantage of an elliptical network with respect to a plane network is obvious here, as, for the same OPO, but with a plane network,

described at line 8 of Table 3, the angle of rotation $\Delta\alpha$ which is also the angle of incidence with respect to the normal to the plane network reaches a maximum value of 36°.

Fig. 12 relates to a device derived from the one in Fig. 11: the non-linear medium is reduced to a portion of an ellipse with an aperture β between 0° and 180° wherein a network is inscribed, the period of which varying from Δ_{max} to Δ_{min}. S_1 and S_2 are the two curved contours on the non-linear medium. The tunability of the device is obtained by rotation of the ellipse portion around point O.

Example 13: <u>Calculation of the variation of the coherence length as a function of the rotation angle in the case of a periodic circular network</u>

Examples 7, 8, 9 and 10 relate to materials for which the index of refraction may vary or not as a function of α.

Fig. 13 relates to a device specific to crystals for which the refractive indexes, n^+ or n^- defined in formula (4) of example 1, vary according to the direction of propagation in a given plane: this is the case in an arbitrary plane of a biaxial crystal or in the main (xz) and (yz) planes of an uniaxial crystal as defined in example 1. The non-linear crystal is machined into a cylinder, the plane of which is one of the planes described earlier so that the refractive index varies continuously from one radius to another.

In the cylinder, a network is inscribed, formed by concentric circular crowns of width p, the periodicity of which $\Delta = 2p$ is constant regardless of the relevant radius, located by angle α. Tunability of the device is however possible by varying the index of refraction as a function of the radius along which the radiations propagate. Thus, the coherence length depends on α. For example, for a parametric interaction with 3 waves of angular frequencies ω_1, ω_2, ω_3, such that $\omega_1 + \omega_2 = \omega_3$, the length of coherence $L_c(\alpha)$ is given by the following relationship:

$$L_c(\alpha) = \frac{\pi c}{\omega_3 n(\omega_3,\alpha) - [\omega_1 n(\omega_1,\alpha) + \omega_2 n(\omega_2,\alpha)]} = \frac{\Delta}{2} = p \quad (22)$$

$n(\omega_i,\alpha)$ is the index of refraction, n^+ or n^-, at ω_i, given by formula (4) of example 1, along the direction of propagation corresponding to a given angle of rotation α.

Thus, to each α corresponds a specific triplet $(\omega_1, \omega_2, \omega_3)$: the tunability of the device is therefore actually obtained by rotation of the cylinder around its center.

For example, a cylinder machined in the (xy) plane of $CsTiOAsO_4$ in which is inscribed a network with period $\Delta = 16\ \mu m$ provides a frequency doubling interaction $(\omega_1 = \omega_2 = \omega, \omega_3 = 2\omega)$. Both waves at fundamental angular frequency ω have an index of

refraction $n(\lambda,\phi)$ such that:

(23) $$n(\lambda,\phi)=\left[\cos^2\phi n_y^{-2}(\lambda)+\sin^2\phi n_x^{-2}(\lambda)\right]^{-1/2}$$

$n_y(\lambda)$ and $n_x(\lambda)$ are given by Sellmeier's equations (8) from Example 1, ϕ is the angle formed by the direction of propagation with the x axis.

The generated wave has an index of refraction $n_z(\lambda)$ given by equation (8) of example 1. Such a structure provides doubling of frequency for wavelengths between 2.56 μm at $\phi = 0°$ and 3.38 μm at $\phi = 90°$.

It is understood that the present invention is not limited to the described and illustrated embodiments above, but it encompasses all the alternatives. Thus notably, the crystal may have a volume of a cylinder 1, a volume of a cylindroid 2, a volume of a truncated cylinder 3 or a volume of a truncated cylindroid 4, or a volume of a portion 5 of a cylinder or cylindroid, as illustrated by Fig. 2. The cylinder or cylindroid may have a section selected from a circular section and an elliptical section.

CLAIMS

1. A device for generating, through interaction(s) with three or four waves from one or more incident optical radiations, one or more emerging optical radiations, at least tunable in frequency, characterized in that it is essentially formed by a crystal with a non-linear optical property, the surface of which defines a cylindrical volume of revolution, in a complete or a truncated way, on at least two opposite and symmetrical quadrants with respect to its axis of revolution, or else even in a partial way on only one of such quadrants, and in that it further comprises an optical system for confining and focussing said incident optical radiation(s) on the central portion(s) of said crystal on the one hand, and for collimating and directing said emerging optical radiation(s) on the other hand.

2. The device according to claim 1, characterized in that said crystal has a volume selected from a cylinder volume (1), a cylindroid

volume (2), a truncated cylinder volume (3), a truncated cylindroid volume (4), a partial cylinder or cylindroid volume (5).

3. The device according to any of the preceding claims, characterized in that said cylindrical volume has a section selected from a circular section and an elliptical section.

4. The device according to any of the preceding claims, characterized in that said crystal includes at least one hyperpolarizable chemical entity.

5. The device according to any of the preceding claims, characterized in that said crystal is a crystal selected from a crystal of $LiTaO_3$, $KTiOPO_4$, $KTiOAsO_4$, $RbTiOPO_4$, $RbTiOAsO_4$, $CsTiOAsO_4$, $\beta.BaBO_4$, LiB_2O_5, $KNbO_3$, $LiIO_3$, $LiNbO_3$, KD_2PO_4, $KH_2P\ O_4$, $NH_4H_2PO_4$, CsD_2ASO_4, CsH_2AsO_4, $AgGaS_2$, $AgGAsE_2$, $ZnGeP_2$, Tl_3AsSe_3 and a crystal of GaAs.

6. The device according to any of the preceding claims, characterized in that the size of said crystal is selected from a micrometric, millimetric size and a centimetric size.

7. The device according to any of the preceding claims, characterized in that said optical system is essentially formed by two components placed on both sides of said crystal and selected from a convergent length, a divergent length, a set of lenses, a reflecting surface or mirror with the concavity orientated on the side of said crystal, and a reflecting surface or mirror with the concavity

orientated on the opposite side of said crystal.

8. The device according to any of the preceding claims, characterized in that the axis of revolution of said crystal coincides with a rotary mechanical axis so that the crystal may rotate around its axis.

9. The device according to any of claims 1 to 8, characterized in that said crystal is a crystal with a phase tuning property through double refraction.

10. The device according to claim 9, characterized in that said crystal is a monocrystalline crystal.

11. The device according to any of claims 1 to 8, characterized in that said crystal is a crystal with a quasi phase tuning property.

12. The device according to claim 11, characterized in that said crystal has, along the direction of propagation of the sought-after radiations, a periodically alternating juxtaposition of monocrystalline domains.

13. The device according to any of the preceding claims, characterized in that the axis of revolution of said crystal is orthogonal to the plane of the direction(s) of wave vectors of an sought-after interaction, and more particularly of the direction(s) providing maximum efficiency of this interaction.

14. The device according to any of the preceding claims, characterized in that said crystal contains direction(s) provide a maximum efficiency of the interaction, so that they are accessible to said

incident optical radiation(s) under normal incidence or close to the normal on one or more surface(s) of said crystal defining a cylindrical volume of revolution, either by rotation of said crystal around its axis of revolution, or by rotation of said incident optical radiations around said crystal in a plane orthogonal to the axis of revolution of said crystal.

15. The device according to any of the preceding claims, characterized in that said incident optical radiation(s) comprise (each) one, two, three or four equal or different frequencies, with colinear or non-colinear wave vectors, and under normal incidence or close to the normal on one or more surfaces of said crystal defining a cylindrical volume of revolution.

16. The device according to any of the preceding claims, characterized in that said crystal has a network of monocrystalline domains selected from a network of plane monocrystalline domains, a network of circular monocrystalline domains, a network of elliptical monocrystalline domains.

17. The device according to any of the preceding claims, characterized in that said crystal has a network of periodically alternating domains, optionally surrounded by a non-alternating monocrystalline crown (c).

18. The device according to any of the preceding claims, characterized in that said incident optical radiations are laser radiation(s), notably one or more laser radiations selected from a fixed frequency laser

radiation and a tunable frequency laser radiation.

19. The device according to any of the preceding claims, characterized in that said interaction(s) are interactions (s) selected from a three-wave interaction or a four-wave interaction.

20. The device according to any of the preceding claims, characterized in that said crystal has a non-centrosymmetric structure so that said device provides a three-wave interaction.

21. The device according to any of the preceding claims, characterized in that said or at least one of said incident optical radiation(s) comprises two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to the sum of said two, or, if required, said three frequencies comprised in said incident optical radiation(s).

22. The device according to any of the preceding claims, characterized in that said or at least one of said emerging optical radiation(s) comprise a frequency which is equal to the double or the triple of a frequency comprised in said or at least one of said incident optical radiation(s).

23. The device according to any of the preceding claims, characterized in that said or at least one of said incident optical radiation(s) comprise two frequencies for a three-wave interaction, or three

frequencies for a four-wave interaction and in that said or at least one of said emerging optical radiation(s) comprise a frequency which corresponds to a difference between said two, or if required, said three frequencies comprised in said incident optical radiation(s).

24. The device according to any of the preceding claims, characterized in that said or at least one of said emerging optical radiation(s) comprise two frequencies for a three-wave interaction, or three frequencies for a four-wave interaction, the sum of which is equal to a frequency comprised in said or at least one of said incident optical radiation(s).

25. The device according to any of the claims 1 to 24, characterized in that said or at least one of said interaction(s) is an interaction with colinear wave vectors.

26. The device according to any of claims 1 to 24, characterized in that said interaction is an interaction with non-colinear wave vectors.

27. The device according to any of the preceding claims, characterized in that said or at least one of said interaction(s) is an interaction selected from an optical parametric amplification, a generation of second or third harmonic.

28. The device according to any of the preceding claims, characterized in that said crystal is placed inside a cavity providing a resonant interaction, and in that said optical system for confining and focussing

said incident optical radiation(s) on the central portion(s) of said crystal on the one hand, and for collimating and directing said emerging optical radiation(s) on the other hand, is placed outside said cavity.

29. The device according to claim 28, characterized in that said resonant interaction is an interaction with three or four waves selected from an optical parametric oscillation, an optical parametric amplification, a generation of second or third harmonics.

30. The device according to claim 28 or 29, characterized in that said cavity includes input and output reflecting surfaces facing each other providing resonance for at least one of the interacting waves.

31. The device according to any claim 30, characterized in that said input reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

32. The device according to claim 30 or 31, characterized in that said at least one resonant wave has a non-zero double refraction angle ρ and in that said output reflecting surface has a radius of curvature, with a concavity selected from a concavity orientated on the side of said crystal and a concavity

orientated on the opposite side of said crystal, so that the outgoing and returning beams coincide.

33. The device according to any of the claims 30-32, characterized in that said at least one resonant wave has a non-zero double refraction angle ρ, and in that said output reflecting surface is placed at a distance (d) from said crystal and has a radius of curvature R the respective values of which satisfy equation $R = d - L$ with d larger than L for a concavity orientated on the side of said crystal, or the equation $R = L - d$ with d less than L for a concavity orientated on the opposite side of said crystal, with L defined as $L = R_c\ (\cos(2\rho) + (\sin(2\rho)\ /\ \tan(\rho_e)) - 1)$, with R_c the radius of the cylindrical volume of revolution, ρ the double refraction angle and with ρ_e defined by $\rho_e = \arcsin(n \sin(2\rho) - 2\rho)$, with n being the refractive index of said at least one wave for which resonance is sought.

34. The device according to claim 30 or 31, characterized in that said at least one resonant wave has a zero double refraction angle ρ, and in that said output reflecting surface is selected from a plane reflecting surface and a reflecting surface having a radius of curvature, with the concavity selected from a concavity orientated on the side of said crystal and a concavity orientated on the opposite side, in order to optimize the oscillation threshold and the stability of the cavity.

35. The device according to any of the preceding claims, characterized in that it further comprises means for thermostatic control of said crystal.

36. The device according to any of the preceding claims, characterized in that said crystal is held at a temperature different from room temperature.

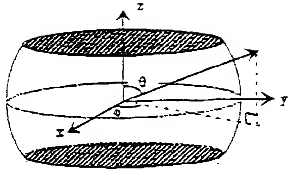
37. The device according to any of the preceding claims, characterized in that it further comprises means for applying a static or low frequency electric field to the inside of said crystal.

38. The device according to any of the preceding claims, characterized in it further comprises a pair of electrodes placed on the opposite faces of said crystal.

39. The device according to any of the preceding claims, characterized in that it forms a component selected from a spectroscope component, a remote detection system component, a remote transmission system component, a remote guiding system component, a LIDAR system component, an optronic counter-measure system component.

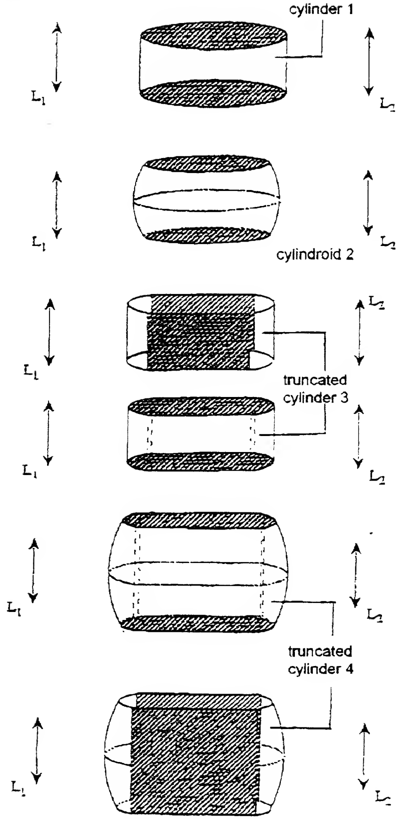
40. A method for generating an optical radiation at least tunable in frequency, characterized in that it implements a device according to any of the preceding claims.

Fig. 1



cylindroid 2

Fig. 2


cylinder 1
L1
L2
cylindroid 2
truncated
cylinder 3
truncated
cylinder 4

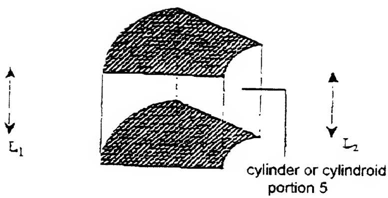


Fig. 2 (continued)

Fig. 3

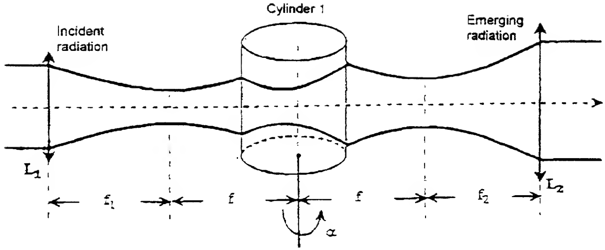

Cylinder 1
Incident radiation
Emerging radiation
L1
L2
f1
f
f
f2
α

Fig. 4

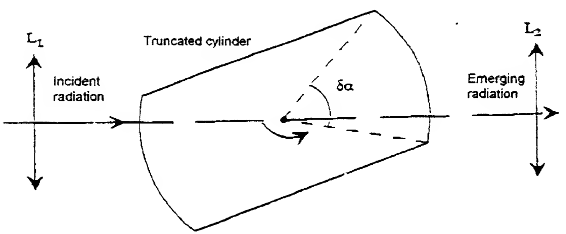
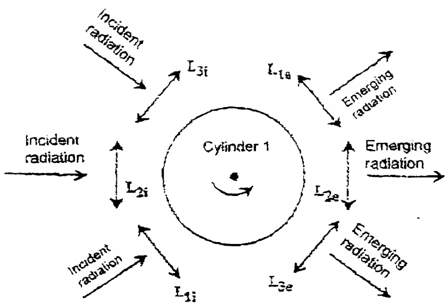

L1
Truncated cylinder
Incident radiation
δα
L2
Emerging radiation

Fig. 5


Incident radiation
L_{3i}
L_{1e}
Emerging radiation
Incident radiation
Cylinder 1
Emerging radiation
L_{2i}
L_{2e}
Incident radiation
L_{1i}
L_{3e}
Emerging radiation

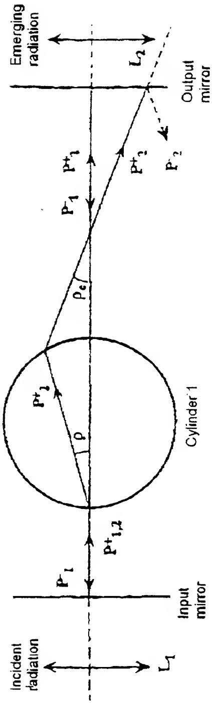

Emerging radiation
L_2
Output mirror
P^{+}_{1}
P^{-}_{1}
P^{+}_{2}
P^{-}_{2}
ρ_e
P^{+}_{2}
ρ
Cylinder 1
$P^{+}_{1,2}$
P^{-}_{1}
Input mirror
Incident radiation
L_1

Fig 6

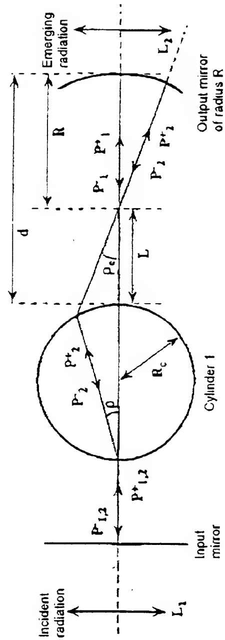

Emerging radiation
L_2
Output mirror of radius R
R
d
L
ρ_e
P^-_1
P^+_1
P^-_2
P^+_2
R_c
ρ
Cylinder 1
$P^-_{1,2}$
$P^+_{1,2}$
Input mirror
Incident radiation
L_1

Fig.7

Fig.8

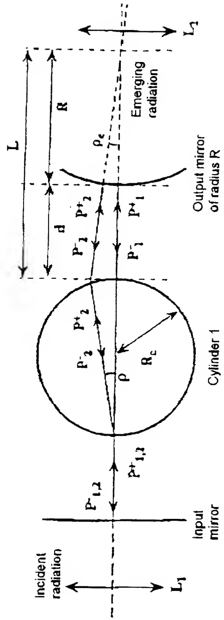

Incident radiation
L1
Input mirror
P-1,2
P+1,2
Cylinder 1
P-2
P+2
ρ
Rc
P-1
P+1
d
L
R
Output mirror of radius R
ρe
Emerging radiation
L2

Fig.9

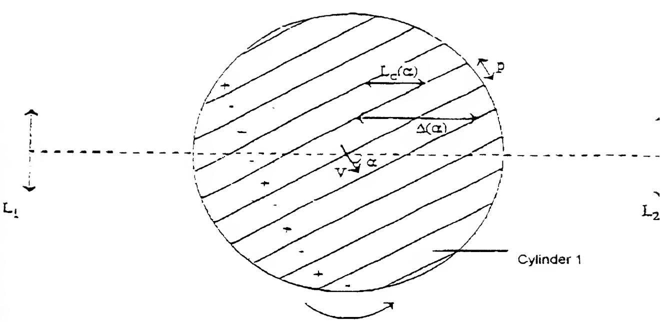

$L_c(\alpha)$
p
$\Delta(\alpha)$
V
α
L_1
L_2
Cylinder 1

Fig.10

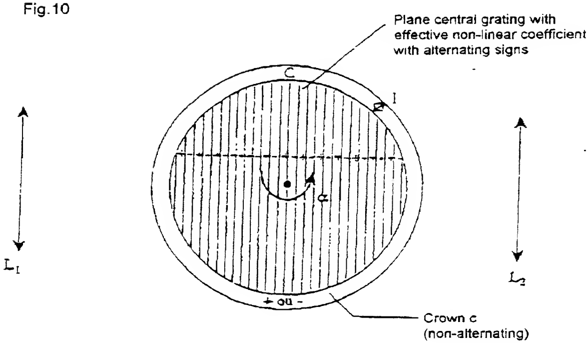

Plane central grating with effective non-linear coefficient with alternating signs
C
I
α
L_1
L_2
+ ou -
Crown c (non-alternating)

Fig. 11

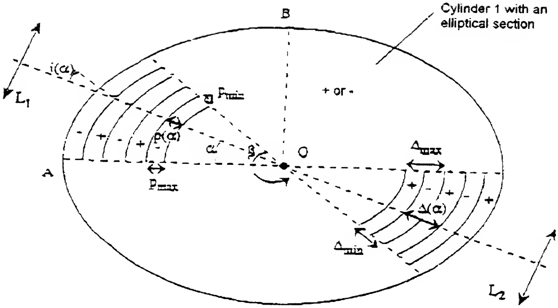

Cylinder 1 with an elliptical section
B
A
O
+ or -
L_1
L_2
$i(\alpha)$
P_{min}
P_{max}
$p(\alpha)$
α
β
Δ_{max}
$\Delta(\alpha)$
Δ_{min}

Fig. 12

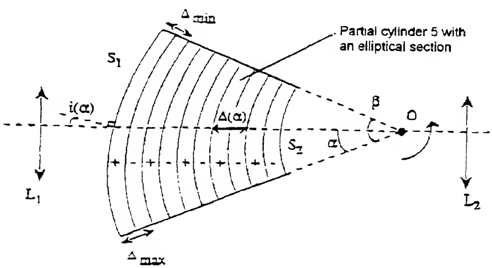
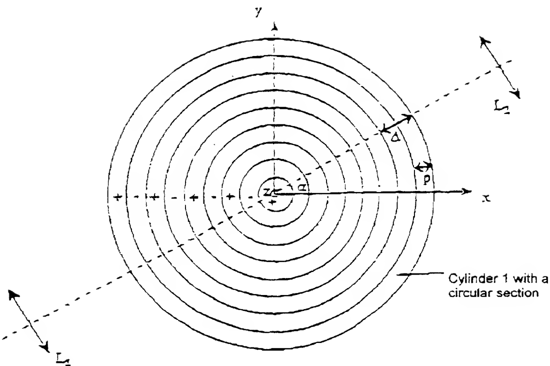

Δ_{min}
Partial cylinder 5 with an elliptical section
S_1
S_2
$i(\alpha)$
$\Delta(\alpha)$
β
O
α
L_1
L_2
Δ_{max}

Fig. 13



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Nixon & Vanderhye P.C. (10/99)
(Domestic Non-Assigned/Foreign) Page 1

RULE 63 (37 C.F.R. 1.63)
INVENTORS DECLARATION FOR PATENT APPLICATION
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

As a below named inventor, I hereby declare that my residence, mailing address and citizenship are as stated below next to my name, and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

MEANS FOR GENERATING OPTICAL RADIATIONS TUNEABLE AT LEAST IN FREQUENCY

the specification of which (**check** applicable **box(s)**):

☐ is attached hereto
☐ was filed on ______ as U.S. Application Serial No. ______ (Atty Dkt. No. 1721-30)
☒ was filed as PCT International application No. PCT/FR98/02563 on 27 November 1998
and (if applicable to U.S. or PCT application) was amended on ______

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose to the Patent Office all information known to me to be material to patentability as defined in 37 C.F.R. 1.56. I hereby claim foreign priority benefits under 35 U.S.C. 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed or, if no priority is claimed, before the filing date of this application:

Priority Foreign Application(s):

Application Number	Country	Day/Month/Year Filed
FR 97/14947	FR	27 November 1997

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

Application Number	Date/Month/Year Filed

I hereby claim the benefit under 35 U.S.C. 120/365 of all prior United States and PCT international applications listed above or below:

Prior U.S./PCT Application(s): Application Serial No.	Day/Month/Year Filed	Status: patented pending, abandoned
PCT/FR98/02563	27 November 1998	Pending

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon. And on behalf of the owner(s) hereof, I hereby appoint **NIXON & VANDERHYE P.C., 1100 North Glebe Rd., 8th Floor, Arlington, VA 22201-4714, telephone number (703) 816-4000 (to whom all communications are to be directed)**, and the following attorneys thereof (of the same address) individually and collectively owner's/owners' attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith and with the resulting patent: Larry S. Nixon, 25640; Arthur R. Crawford, 25327; James T. Hosmer, 30184; Robert W. Faris, 31352; Richard G. Besha, 22770; Mark E. Nusbaum, 32348; Michael J. Keenan, 32106; Bryan H. Davidson, 30251; Stanley C. Spooner, 27393; Leonard C. Mitchard, 29009; Duane M. Byers, 33363; Jeffry H. Nelson, 30481; John R. Lastova, 33149; H. Warren Burnam,
29 Jr. 29366; Mary J. Wilson, 32955; J. Scott Davidson, 33489; Alan M. Kagen, 36178; Robert A. Molan, 29834; B. J. Sadoff, 36663; James D. Berquist, 34776; Updeep S. Gill, 37334; Michael J. Shea, 34725; Donald L. Jackson, 41090; Michelle N. Lester, 32331; Frank P. Presta, 19828; Joseph S. Presta, 35329; Joseph A. Rhoa, 37515; Raymond Y. Mah, 41426; Chris Comuntzis, 31097. I also authorize Nixon & Vanderhye to delete any attorney names/numbers no longer with the firm and to act and rely solely on instructions directly communicated from the person, assignee, attorney, firm, or other organization sending instructions to Nixon & Vanderhye on behalf of the owner(s).

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☒ *See attached sheet(s) for additional inventor(s) information!!*

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Inventor: (first) MI (last) (citizenship)
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FOR ADDITIONAL INVENTORS, check box ☐ **and attach sheet with same information and signature and date for each.**